An unknown source of reactor radionuclides in the Baltic Sea revealed by multi-isotope fingerprints

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We present an application of multi-isotopic fingerprints (i.e., 236U/238U, 233U/236U, 236U/129I and 129I/127I) for the discovery of previously unrecognized sources of anthropogenic radioactivity. Our data indicate a source of reactor 236U in the Baltic Sea in addition to inputs from the two European reprocessing plants and global fallout. This additional reactor 236U may come from unreported discharges from Swedish nuclear research facilities as supported by high 236U levels in sediment nearby Studsvik, or from accidental leakages of spent nuclear fuel disposed on the Baltic seafloor, either reported or unreported. Such leakages would indicate problems with the radiological safety of seafloor disposal, and may be accompanied by releases of other radionuclides. The results demonstrate the high sensitivity of multi-isotopic tracer systems, especially the 233U/236U signature, to distinguish environmental emissions of unrevealed radioactive releases for nuclear safeguards, emergency preparedness and environmental tracer studies.

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$^{236}\text{U}$ ($t_{1/2} = 2.34 \times 10^7$ years) is an isotope of uranium that is produced by thermal neutron capture of $^{235}\text{U}$ via (n, γ)-reactions and through $^{238}\text{U}$ (n, 3n) $^{236}\text{U}$ reactions with fast neutrons. Even though a small amount of $^{236}\text{U}$ (~35 kg) occurs naturally in the Earth’s crust, $^{236}\text{U}$ is (by mass) the largest secondary product created in nuclear reactors, estimated to be $\sim 10^6$ kg. $^{236}\text{U}$ is therefore a sensitive tracer of deliberate or accidental leakage from the nuclear fuel/waste cycle. The known sources of reactor $^{236}\text{U}$, i.e., deliberate releases from the two European reprocessing plants at La Hague, France (LH), and Sellafield, UK (SF) since 1950s, can be traced throughout the North Atlantic and the Arctic water currents. Emissions from other known sources of reactor $^{236}\text{U}$, e.g., the Springfield nuclear facility and the Fukushima accident, are negligible.

A significant amount of $^{236}\text{U}$ (estimated at >1000 kg) was also delivered to the Earth’s surface environments from the global fallout of atmospheric nuclear weapons testing in the 1950s and 1960s. This ubiquitous fallout signature can make identification of sources of reactor $^{236}\text{U}$ challenging because of methodological difficulties in distinguishing the source of $^{236}\text{U}$. In addition, the $^{236}\text{U}/^{238}\text{U}$ ratio does not provide source information because of the prevalence of $^{238}\text{U}$ in nature.

Reactor $^{236}\text{U}$ can be differentiated from fallout $^{236}\text{U}$ because these sources have different and characteristic $^{233}\text{U}/^{236}\text{U}$ ratios due to different nuclear production mechanisms. $^{233}\text{U}$ was mostly produced during nuclear weapons testing by fast neutrons via $^{235}\text{U}(n, 3n)^{233}\text{U}$ reactions or directly by $^{233}\text{U}$-fueled devices, whereas almost no $^{233}\text{U}$ is produced in thermal nuclear power reactors or reprocessing plants. Recently, $^{233}\text{U}$ measurements at environmental levels have become possible with advanced accelerator mass spectrometry.

The representative $^{233}\text{U}/^{236}\text{U}$ atomic ratio of global fallout from atmospheric nuclear weapons testing was suggested to be $(1.40 \pm 0.15) \times 10^{-2}$. This is several orders of magnitude higher than the $^{233}\text{U}/^{236}\text{U}$ atomic ratio in nuclear reactors, e.g., $1 \times 10^{-7} - 1 \times 10^{-6}$ in LH discharges, which agrees well with reactor model calculations. In the Irish Sea, an average $^{233}\text{U}/^{236}\text{U}$ atomic ratio of $(0.12 \pm 0.01) \times 10^{-2}$ has been measured, reflecting a dominant reactor signal released from SF. The use of the $^{233}\text{U}/^{236}\text{U}$ atomic ratio helps to deconvolve the origin of $^{236}\text{U}$ based on the characteristic $^{233}\text{U}/^{236}\text{U}$ fingerprint from different source terms. In addition, the combination of $^{236}\text{U}$ with other radionuclides, e.g., $^{129}\text{I}$, can be useful to trace the transport of $^{236}\text{U}$ from specific source points, e.g., releases from LH and SF.

The Baltic Sea is a highly polluted sea, with anthropogenic radionuclides demanding specific attention because of the risk to ecosystem and humans from radioactivity in the environment. It receives radionuclides from global fallout, discharges from the two European reprocessing plants, releases from the Chernobyl accident, and from any other local sources. In this study, we use a novel combination of three anthropogenic radionuclides—$^{233}\text{U}$, $^{236}\text{U}$, and $^{129}\text{I}$—to identify a previously unknown local source of radionuclide pollution to the Baltic Sea.

**Results and discussion**

**Study area and sampling.** The Baltic Sea is a landlocked, intracranial sea in Northern Europe with about 80 million inhabitants in the surrounding states and constitutes one of the largest brackish water environments on Earth. The water exchange of this large brackish estuarine-like water mass with the Kattegat and the North Sea takes place through the narrow and shallow Danish Straits (Fig. 1). The driving force for the water circulation is freshwater surplus from river runoff, estimated at 473 km$^3$ per year, together with “recycled” North Sea inflowing water as Baltic outflow that sum to a total water exchange rate of 753 km$^3$ per year. A mean residence time for the 21,721 km$^3$ Baltic water volume was estimated to be 29 years, which is equivalent to a “half-life” for the water volume of 20 years.

In the investigation presented here, water and sediment samples were collected from the Baltic Sea and related water masses including the western Danish coast, from 2011 to 2016 (Supplementary Tables 1 and 2). The majority of water samples are from the surface (0–5 m depth), with a few samples from deep water, and one lake water from the Lake Mälaren, which receives downstream discharges from a nuclear fuel fabrication facility (Westinghouse) in Sweden and finally drains into the Baltic Sea.

In addition to the Baltic Sea water, we analyzed sediment samples to assess the accumulation trend of the isotopes in the Baltic Sea. A more detailed description of the study area and samples can be found in the “Methods” section.

To facilitate the presentation of results and related discussion, we grouped the sampling locations into five geographical regions (Fig. 1) in the Baltic Sea including (1) KGR: Kattegat–Skagerrak region including the Kattegat, Skagerrak and Danish west coast nearby the North Sea; (2) DS: Danish Straits including the Belt Seas and the Sound; (3) SBR: South Baltic Sea region including Arkona Basin, Bothnian Basin, and South Baltic Proper; (4) MBR: Middle Baltic Sea region including Northern Baltic Proper, Western Gotland Basin, Eastern Gotland Basin, and Gulf of Riga; and (5) NBR: North Baltic Sea region including Archipelago and Åland Sea, Bothnian Sea and Bothnian Bay.

**Spatial pattern of $^{236}\text{U}$ concentration and $^{236}\text{U}/^{238}\text{U}$ and $^{233}\text{U}/^{236}\text{U}$ atomic ratios.** The measured $^{236}\text{U}/^{238}\text{U}$ atomic ratios (Supplementary Tables 1 and 2) vary within $(5–52) \times 10^{-9}$, with the higher ratios in the central and northern parts of the Baltic Sea and lower ratios in the western parts (Danish Straits, Kattegat, Skagerrak, and Danish west coast). The highest value reported here is sixfold greater than the average value found in the North Sea in 2010 $(7.6 \pm 3.7) \times 10^{-9}$. The spatial patterns (Fig. 2) suggest a general decline of $^{236}\text{U}$ with distance from higher values in the North Sea which is expected to be dominated by discharges from LH and SF. However, high $^{236}\text{U}$ concentrations $(6–9) \times 10^{7}$ atom/l are observed in the surface water of the Bothnian Sea and Bothnian Bay, which are comparable to values $(5–10) \times 10^{7}$ atom/l in the central North Sea. Compared to the Kattegat–Skagerrak region, the average $^{236}\text{U}/^{238}\text{U}$ atomic ratio in the middle and north Baltic region increases by a factor of 3, from $(10 \pm 3) \times 10^{-9}$ to $(32 \pm 7) \times 10^{-9}$. This pattern of increasing $^{236}\text{U}/^{238}\text{U}$ ratio highlights an additional, likely local, source of $^{236}\text{U}$ in the Baltic Sea.

$^{233}\text{U}/^{236}\text{U}$ atomic ratios obtained here are in the range of $(0.14–0.87) \times 10^{-2}$, with the lowest $^{233}\text{U}/^{236}\text{U}$ atomic ratios in the western parts of the Baltic, including the Danish coast, and the highest ratios in the central Baltic Sea. As the typical $^{233}\text{U}/^{236}\text{U}$ ratio for global fallout is $(1.4 \pm 0.1) \times 10^{-2}$, the high $^{233}\text{U}/^{236}\text{U}$ in the central Baltic Sea could indicate either strong influence of global fallout or addition from a local source.

**Distribution of $^{129}\text{I}$ concentration, $^{129}\text{I}/^{127}\text{I}$ and $^{236}\text{U}/^{129}\text{I}$ atomic ratios.** The measured $^{129}\text{I}$ concentrations $(3–232) \times 10^{9}$ atom/l and $^{129}\text{I}/^{127}\text{I}$ atomic ratios $(101–1286) \times 10^{-9}$ in the seawater collected in this work show comparable values and distribution trends as observed in an earlier investigation, with the highest values in the North Sea-Skagerrak–Kattegat, decreasing values toward the Sound and relatively constant values in the Baltic Proper. The distributions of $^{129}\text{I}$ concentrations and $^{129}\text{I}/^{127}\text{I}$ atomic ratios indicate that the major source of $^{129}\text{I}$ in the
Baltic Sea are marine discharges from the two nuclear reprocessing plants at LH and SF. The water mass pathways from these plants have been shown to contain appreciable amounts of $^{129}$I along the passage to the Baltic Sea\(^4\).

Aldahan et al.\(^22\) reported that the average concentration of $^{129}$I in the rivers around the Baltic Sea was 3.9 x 10^8 atom/l, which suggested some minor contribution of $^{129}$I from riverine water to the Baltic Sea. The $^{129}$I concentrations obtained in this work show a larger gradient (two orders of magnitude) compared to the $^{238}$U concentrations (15-fold) along the Baltic Sea. $^{236}$U/$^{129}$I ratios are within the range of (5 – 133) x 10^{-4} and indicate a reversed geographical distribution compared to $^{129}$I concentration and $^{129}$I/$^{127}$I atomic ratio (Fig. 2).

**Potential sources of uranium and iodine in the Baltic Sea.** Five different sources of uranium and iodine in the Baltic Sea are:

1. Natural ocean water, with salinity of 35‰, which contains ~60 μg/l $^{127}$I, 3 μg/l $^{238}$U, but negligible $^{239}$U, $^{236}$U, and $^{233}$U.
2. Natural freshwater with salinity <1‰, negligible $^{129}$I, $^{236}$U, and $^{233}$U, and significantly lower $^{127}$I and $^{238}$U than seawater (0.05–10 μg/l for both nuclides).
3. Global fallout from atmospheric nuclear weapons testing, with negligible $^{127}$I and $^{238}$U, an average $^{233}$U/$^{236}$U atomic ratio of (1.40 ± 0.15) x 10^{-2}, and a surface geographical distribution pattern for $^{236}$U and $^{233}$U similar to that of other actinides (e.g., Pu) from global fallout\(^23\). Earlier studies have estimated $^{236}$U concentration (up to 1.4 x 10^8 atom/l peaking in 1960s) in surface water of the North Sea to be related to global fallout, which might have been partly masked by discharges from the nuclear reprocessing of LH and SF\(^{24,25}\). In the Baltic Sea, with an average depth of 55 m, the dilution by vertical dispersion is limited, and a ten times higher concentration is expected for the same inventory, which might mimic higher input. The $^{233}$U/$^{236}$U atomic ratio of the global fallout contribution is expected to be constant after 1980 when all countries stopped aboveground nuclear bomb tests. Concentration of $^{236}$U in river runoff is expected to have reduced over the decades, while the $^{233}$U/$^{236}$U atomic ratio stays constant.
4. Marine discharges from European nuclear fuel reprocessing plants (including mainly SF and LH), with known $^{236}$U and $^{129}$I source functions\(^24,26\), but negligible amounts of $^{127}$I and $^{238}$U. This source dominates the $^{236}$U and $^{129}$I budget of marine water entering the Skagerrak from the North Sea. Compared to $^{236}$U, almost no $^{233}$U is produced in thermal nuclear reactors, and thus $^{236}$U should also be absent from marine discharges of the reprocessing plants.
5. The Chernobyl accident. Pu from Chernobyl has been found in fallout over central Europe\(^27\) and, as Pu and U are refractory elements transported similarly by atmospheric dispersion, Chernobyl $^{236}$U should have been deposited following a similar pattern as Pu isotopes. Consequently, a Chernobyl signal of $^{236}$U may be present in river runoff and marine waters. Based on the present understanding of the production mechanisms of $^{233}$U, it is expected that Chernobyl fallout is not a significant contributor of $^{233}$U in this context.

Waters entering the Baltic Sea from the North Sea have $^{236}$U/$^{238}$U and $^{233}$U/$^{236}$U atomic ratios set by the balance of reprocessing discharge and global fallout\(^9,20\). As they are distributed in the Baltic and mix with waters from various rivers, ratios can be altered by addition from local sources of $^{236}$U and $^{233}$U (and minor $^{238}$U in
river waters). Removal of uranium from Baltic water will not alter the ratios. The increase in $^{236}\text{U}/^{238}\text{U}$ observed within the Baltic Sea points clearly to a local source of this anthropogenic radionuclide.

$^{236}\text{U}$ source identification via binary mixing. The concentration of $^{238}\text{U}$ (Fig. 3A) demonstrates a strong positive correlation ($R^2 = 0.91$) with salinity. The intercept corresponds to the average riverine input with a $^{238}\text{U}$ concentration of $0.33 \pm 0.05 \mu g/l$, which falls in the range (0.2–0.7 $\mu g/l$) of $^{238}\text{U}$ for some rivers in the Baltic Sea region. We will use the typical value $0.4 \mu g/l$ in the following calculations. There is more scatter in the $^{238}\text{U}$ concentration for low salinities, which might be attributed to differences in regional riverine input. $^{129}\text{I}$ also shows a general positive linear correlation with salinity demonstrated by two mixing lines for the western (KGR-DS, $R^2 = 0.89$) and interior (SBR-MBR-NBR, $R^2 = 0.97$) region (Fig. 3B). The scatter at the high salinity end can be attributed to the mixing of $^{129}\text{I}$ enriched North Sea coast water with $^{129}\text{I}$ depleted North Atlantic water in the Kattegat–Skagerrak region. The $^{238}\text{U}$ and $^{129}\text{I}$ trends with salinity suggest that their concentrations in the Baltic Sea are mainly controlled by the saline water input from the North Sea via Kattegat–Skagerrak, mixing with fresh waters in the basin.

Both the $^{236}\text{U}/^{238}\text{U}$ and $^{236}\text{U}/^{129}\text{I}$ atomic ratios increase with the decreasing salinity as waters mix in the interior of the Baltic Sea. The $^{236}\text{U}/^{238}\text{U}$ ratio increases by a factor of 3, while the $^{236}\text{U}/^{129}\text{I}$ ratio increases greater than an order of magnitude from an average of $(8 \pm 2) \times 10^{-4}$ in the Kattegat–Skagerrak region, corresponding to reprocessing derived $^{236}\text{U}$ and $^{129}\text{I}$, to $1 \times 10^{-2}$ in the central Baltic Sea. Both ratios indicate addition of $^{236}\text{U}$ from a local source. If the source does not contain any $^{129}\text{I}$, the

![Fig. 2 Results of anthropogenic radionuclides. Distribution of $^{236}\text{U}$ and $^{129}\text{I}$ concentrations, and $^{236}\text{U}/^{238}\text{U}$, $^{129}\text{I}/^{127}\text{I}$, $^{233}\text{U}/^{236}\text{U}$, and $^{236}\text{U}/^{129}\text{I}$ atomic ratios in the Baltic Sea surface water during 2011–2016.](https://doi.org/10.1038/s41467-021-21059-w)
A tenfold increase in $^{236}\text{U}/^{129}\text{I}$ suggests that ca. 90% of $^{236}\text{U}$ in the central Baltic Sea is from local sources. If the source does contain $^{129}\text{I}$, the portion of $^{236}\text{U}$ derived locally must be still larger.

To understand the source terms of $^{236}\text{U}$ in the Baltic Sea, a binary mixing model is applied with two respective end members representing $^{236}\text{U}$ input from the North Sea and freshwater input via river runoff. Parameters for the first end member representing the North Sea water entering from the west Baltic Sea are well defined by previous studies (Supplementary Table 3)\textsuperscript{20,29}. The deviation of the observed $^{236}\text{U}/^{238}\text{U}$ atomic ratio in the binary mixing (line L1, Fig. 4A) of the North Sea water and an assumed freshwater end member containing no $^{236}\text{U}$ (neither $^{233}\text{U}$) from the best-fit model L reflects additional $^{236}\text{U}$ sources besides North Sea water. The spatial distribution of deviations in the $^{236}\text{U}/^{238}\text{U}$ atomic ratio enable determination of the location of the additional $^{236}\text{U}$ source (Supplementary Fig. 2). The distribution pattern is compatible with the introduction of additional riverine $^{236}\text{U}$ input from the north Baltic region, which has most river runoff. Nevertheless, it is challenging to define the $^{236}\text{U}/^{238}\text{U}$ ratio of the riverine input to the Baltic because a component of global fallout may still be present in runoff from the land surface. The $^{236}\text{U}/^{238}\text{U}$ and $^{236}\text{U}/^{129}\text{I}$ ratios cannot be used to determine the extent to which the excess $^{236}\text{U}$ is from global fallout or an...
Additional, previously undiscovered, source that has directly released $^{236}$U to the Baltic Sea.

**Application of $^{233}$U/$^{236}$U atomic ratio for $^{236}$U source identification.** If we assume that the excess $^{236}$U originates only from global fallout, the $^{236}$U/$^{238}$U atomic ratio of the riverine input in the best-fit binary mixing is $(6.79 \pm 0.75) \times 10^{-8}$ (line L, Fig. 4A). However, there is a clear deviation of the observation from the model for $^{233}$U/$^{236}$U atomic ratios (Fig. 5A). A subgroup of samples from the Kattegat–Skagerrak reveal a relatively stable $^{233}$U/$^{236}$U atomic ratio of $0.20 \times 10^{-2}$ (blue dash-dotted line in Fig. 5) independent of $^{236}$U/$^{238}$U and salinity. This behavior can be explained by assuming an end member of North Sea water with $^{233}$U/$^{236}$U atomic ratio $=0.20 \times 10^{-2}$ (a mixed signal of global fallout plus nuclear reprocessing) and salinity $=35\%_o$, which is mixed with natural uranium or water with neither $^{236}$U nor $^{233}$U. This feature shows the notable impact of nuclear reprocessing from SF and LH in the region.

On the other hand, a cluster of samples with the majority from the south, middle and north Baltic Sea region, representative for a large part of the Baltic surface water and with median salinity $(6.92 \pm 0.29)\%_o$, show a typical $^{233}$U/$^{236}$U atomic ratio of $(0.53 \pm 0.03) \times 10^{-2}$ (the green dash-dotted line in Fig. 5). This cluster lies significantly below the binary mixing model L, indicating an additional local $^{236}$U source besides the global fallout, which is characterized by low $^{233}$U/$^{236}$U atomic ratio. A low $^{233}$U/$^{236}$U atomic ratio is typical for releases from nuclear reactors, thereby we assume such a reactor-related source of $^{236}$U with negligible $^{233}$U in the following.

About two-thirds of the anthropogenic uranium observed in the middle and north Baltic Sea region seems to originate from this additional local source (Eq. (1)), indicating a strong contribution of $^{236}$U without $^{233}$U, i.e., from a thermal nuclear reactor $^{236}$U.

![Fig. 5 Results of $^{233}$U/$^{236}$U, $^{233}$U/$^{236}$U atomic ratio vs. $^{238}$U/$^{236}$U atomic ratio (A) and salinity (B). KGR Kattegat-Skagerrak region including the Kattegat, Skagerack and Danish west coast nearby the North Sea, DS Danish Straits including the Belt Sea and the Sound, SBR South Baltic Sea region including Arkona Basin, Borholm Basin, and South Baltic Proper, MBR Middle Baltic Sea region including Northern Baltic Proper, Western Gotland Basin, Eastern Gotland Basin and Gulf of Riga, NBR North Baltic Sea region including Archipelago and Åland Sea, Bothnian Sea, and Bothnian Bay, L (blue solid line) the best-fit binary mixing line between the North Sea water and a freshwater end member with salinity $=0$, $^{238}$U$=0.4\mu g/l$, $^{236}$U$=(3.56 \pm 0.39) \times 10^{-10}$ atom/l, which is calculated to match the $^{233}$U/$^{236}$U atomic ratio.](image)

\[
\frac{\text{R}_0}{\text{R}_f} = \frac{N_{233,r} + N_{233,f}}{N_{236,r} + N_{236,f}} = \frac{N_{236,r} \cdot R_f + N_{236,f} \cdot R_f}{N_{236,r} + N_{236,f}} = \frac{R_f + N_{236,f} / N_{236,f} - R_f}{1 + N_{236,f} / N_{236,f}}
\]

where $R_0$, $R_f$, and $R_t$ represent, respectively, the $^{233}$U/$^{236}$U atomic ratio of the Baltic seawater, global fallout, and nuclear reactor; $N_{233,r}$ and $N_{233,f}$ refer to the atomic number of $^{233}$U from global fallout and nuclear rector, respectively; $N_{236,r}$ and $N_{236,f}$ refer to the atomic number of $^{236}$U from global fallout and nuclear reactor, respectively. Therefore, $N_{236,f} = N_{236,r}$ with $R_f = (0.53 \pm 0.03) \times 10^{-2}$, $R_t = 1.4 \times 10^{-2}$, and $R_r = 0.12 \times 10^{-2}$ (the Irish Sea ratio), we calculate the $^{236}$U contribution from our assumed reactor source to be $2.1 \pm 0.2$ times that of global fallout. To locate this additional reactor $^{236}$U source, we apply another binary mixing line L2 (Fig. 4A) of the North Sea water with riverine water, the latter carrying global fallout that accounts for $(1/(1 + 1))$ of the average $^{236}$U concentration of our samples in the Baltic Sea. Thus, the freshwater end member is characterized by salinity $=0$, $^{238}$U$=0.4\mu g/l$, $^{236}$U$=(3.56 \pm 0.39) \times 10^{-10}$ atom/l, which is calculated to match the $^{233}$U/$^{238}$U atomic ratio of $(1.70 \pm 0.18) \times 10^{-9}$ for the cluster of samples from SBR, MBR, and NBR at the media salinity of $(6.92 \pm 0.29)\%_o$ (Supplementary Fig. 2). The resultant $^{236}$U/$^{238}$U atomic ratio of the freshwater end member is $(3.52 \pm 0.39) \times 10^{-8}$. The excesses of $^{236}$U/$^{238}$U atomic ratio from the mixing curve L2 and their spatial distribution are shown in Fig. 6. The data indicate that the extra reactor $^{236}$U source input is not from places where salinity is particularly low or where there are rivers, but in the middle and north basins of the Baltic Sea which is probably linked to direct releases of $^{236}$U into these locations.

**Properties of the unknown $^{236}$U source.** To narrow down the possible sources of the excess $^{236}$U, $^{236}$U inventories and fluxes need to be estimated. It should be noted this calculation is a first-order approximation based only on our data on surface waters from a multi-year survey. A precise interpretation will require more data, and to account for many different effects such as vertical distribution of $^{236}$U in the Baltic water columns, interannual variation in distribution pattern and on the scavenging of uranium into the sediment (especially in the anoxic regions).

The existence of an additional source of anthropogenic $^{236}$U in the Baltic Sea is indicated by the difference between the models L2 and L (Fig. 6). The amount of $^{236}$U required to explain this difference can be calculated by the following approximation and
Fig. 6 Deviation of $^{236}\text{U}/^{238}\text{U}$ from L2. Deviations of $^{236}\text{U}/^{238}\text{U}$ atomic ratio from binary mixing line L2 (A) and their respective geographical distribution on the map (B). KGR Kattegat-Skagerrak region including the Kattegat, Skagerrak and Danish west coast nearby the North Sea, DS Danish Straits including the Belt Seas and the Sound, SBR South Baltic Sea region including Arkona Basin, Borholm Basin, and South Baltic Proper, MBR Middle Baltic Sea region including Northern Baltic Proper, Western Gotland Basin, Eastern Gotland Basin and Gulf of Riga, NBR North Baltic Sea region including Archipelago and Åland Sea, Bothnian Sea, and Bothnian Bay. Average salinity $S = 7.36\%$. Uncertainties are expanded uncertainties using a coverage factor of $k = 1$.

with the uncertainty estimated according to Müller:  

$$X_{236} = \int_0^{35} (L(S) - L_2(S)) \left[\frac{^{238}\text{U}(S)}{^{236}\text{U}}\right] \frac{dV(S)}{dS} \, dS$$

$$\approx (L - L_2)(S) \left[\frac{^{238}\text{U}}{^{236}\text{U}}\right] V_{BS} = 200 \pm 47 \text{g}$$

where $X_{236}$ is the excess mass of $^{236}\text{U}$ in the Baltic Sea, $S$ is the salinity, $\left[\frac{^{238}\text{U}(S)}{^{236}\text{U}}\right]$ is the $^{238}\text{U}$ concentration corresponding to $S$ taken from Fig. 3A. $S$ is the average salinity of the Baltic Sea. $S$ was taken as $7.36\%$ based on the reported mean salinity of the Baltic Sea during 1902–1998. Our data from SBR, MBR, and NBR, which comprise the bulk of Baltic Sea water, show an average salinity of $7.06\%$ and a median of $6.92\%$, comparable to the reported value. $\left[\frac{^{238}\text{U}(S)}{^{236}\text{U}}\right]$ is $0.9\mu g/l$. $V_{BS}$ is the volume of the Baltic Sea ($21,721 \text{km}^3$) and $(L - L_2)(S) = (1.02 \pm 0.24) \times 10^{-8}$ is the difference of the model curves at the average salinity. The approximation in the formula is possible because in the models L and L2, the $^{236}\text{U}$ concentration is a linear function of $S$. Therefore, $200 \pm 47$ g of $^{236}\text{U}$ is from the additional reactor source.

This calculation is a snapshot in time based on the uranium isotope ratios and salinity. While uranium concentrations in water may be altered in the partly anoxic Baltic Sea by precipitation of inorganic U(IV) or binding to organics in the sediment, the uranium isotopic ratios will only change by mixing of different sources. Total salinity is slightly affected by precipitation (rain and snow) and evaporation (net balance $63 \text{ km}^3$ per year)\(^{32}\), which may be neglected at the present level of precision. Large intrusions of the North Sea water can change salinity patterns and introduce anthropogenic uranium from the North Sea. These intrusions add up to $5.2 \times 10^9$ metric tons of salt\(^{32}\), which is about $3\%$ of the salt inventory of the Baltic\(^{32}\). The spatial pattern may not be constant throughout a multi-year survey, nevertheless, a minor change in the calculation is expected as we use only the average salinity for our estimate.

Taking into account that the ratio between the additional source and global fallout is $N_{236,U}/N_{238,U} = 2.1$, it suggests that $95 \pm 22$ g of $^{236}\text{U}$ is related to global fallout introduced into the Baltic Sea directly or via riverine input. It is estimated that a total inventory of $1000 \text{ kg}$ of anthropogenic $^{236}\text{U}$ was distributed via global fallout mainly on the Northern Hemisphere\(^{33}\). Considering the surface area of the Baltic Sea of $3.77 \times 10^8 \text{ km}^2$ (without the catchment area) in comparison to the Northern Hemisphere (i.e., $2.55 \times 10^8 \text{ km}^2$), the total $^{236}\text{U}$ deposition from direct global fallout is estimated as $1.5 \text{ kg}$. However, if considering the 29-year mean residence time (equivalent to 20-year half-life) of Baltic seawater, then most of the deposited $1.5 \text{ kg}$ $^{236}\text{U}$ was transported out of the region after 60 years (i.e., three half-lives), leaving behind $\sim 0.19 \text{ kg}$. In addition, some $^{236}\text{U}$ fraction from global fallout might be removed from the water body and incorporated into the Baltic sediment\(^{34}\). Therefore, the above estimation of $95 \pm 22$ g remaining $^{236}\text{U}$ in the Baltic seawater from global fallout seems plausible, considering the uneven distribution of global fallout.

If we include the Baltic catchment area ($1.64 \times 10^6 \text{ km}^2$) in the calculation, the input of global fallout $^{236}\text{U}$ in the Baltic region can be up to $8 \text{ kg}$ ($1.5 \text{ kg}$ in seawater + $6.4 \text{ kg}$ in catchment area). However, only a small fraction of the particle associated $^{236}\text{U}$ deposited on land can be leached and transported to the Baltic Sea through river runoff. If we assume this fraction accounts for $10\%$ of the $6.4 \text{ kg}$ of $^{236}\text{U}$ deposited in the catchment, the total amount of global fallout $^{236}\text{U}$ in the Baltic Sea might be about $0.64 + 1.5 = 2.14 \text{ kg}$.

Emissions from the Chernobyl accident may contribute additional $^{236}\text{U}$ to the Baltic Sea, but it is difficult to quantify. Nuclear dumping and/or nuclear installations around the Baltic countries are also possible source candidates. As marked in Fig. 1, there are many nuclear installations in surrounding Baltic countries, but there is very limited documentation with poor, unreleased or missing data about the $^{233}\text{U}$ and $^{236}\text{U}$ release records from these installations (Supplementary Table 4)\(^{11}\). Data for $^{236}\text{U}$ are available from Westinghouse during 1998–2017, with a total reported release of $1.06 \times 10^8 \text{ Bq}$ of $^{236}\text{U}$, equal to $0.44 \text{ g}$. In addition, we measured one lake water sample collected in Lake Mälaren (Supplementary Table 2), which receives waste discharges from the Westinghouse facility and finally drains into the Baltic Sea. The results show that the $^{236}\text{U}/^{238}\text{U}$ ratios is at the level of $2 \times 10^{-8}$, which is comparable with the seawater samples collected in the central Baltic Sea. The lake water shows a $^{233}\text{U}/^{236}\text{U}$ atomic ratio of $(0.18 \pm 0.05) \times 10^{-2}$, a signature of reactor material.

The amount of $^{236}\text{U}$ released from the Westinghouse installation ($0.44 \text{ g}$) is negligible compared to the above estimated $280 \text{ g}$ of the unknown reactor source in the Baltic Sea. For the Lake Mälaren, the $^{236}\text{U}$ concentration was measured to be $1.5 \pm 0.1 \mu g/l$ in this work, together with a flux of $166 \text{ m}^3/\text{s}$, it means an input of $0.1 \text{ g}$ per year of $^{236}\text{U}$, which is negligible also.

Another candidate for the additional source may be reactor fuel, dumped into the Baltic. The atomic ratio of $^{236}\text{U}/^{238}\text{U}$ can
be as high as $1 \times 10^{-2}$ in conventional nuclear reactors, which would require only 27 kg of dumped/dissolved fuel (a commercial nuclear reactor contains $\sim 100,000$ kg of fuel). $235\text{U}$ enrichment in reactor fuel is 3% for light-water reactors, up to 10% for thermal gas-cool reactors and up to 20% for fast reactors.\textsuperscript{39} The concentration will be even higher in the core of a nuclear reactor for marine applications, where enriched or highly enriched $233\text{U}$ is used; Russian submarine reactors were reported to contain 50–200 kg of $233\text{U}$.\textsuperscript{36} The former Soviet Union (USSR) was accused of dumping radioactive waste in the Baltic Sea, but it is not possible to assess the dumped amount.\textsuperscript{37,38}

The geographical distribution of $238\text{U}/235\text{U}$ atomic ratio in seawater of the Baltic Sea shows high values nearby the Swedish coast close to Stockholm, which is within 100 km of a nuclear research company Studsvik AB, Nykoping that has been in operation since 1950s. It was reported that during 1959 and 1961, 64 tons of radioactive waste with total radioactivity of 14.8 GBq were dumped into the coastal area nearby Studsvik.\textsuperscript{39} The aquatic discharges of radionuclides (except $3\text{H}$) from Studsvik into the Baltic Sea in 1999–2010 were reported to be 0.45 TBq with the majority consisting of $\text{Sr}$, $137\text{Cs}$, $60\text{Co}$, and $134\text{Cs}$.\textsuperscript{40} Our measurements on some sediment samples from the Studsvik area show very high $236\text{U}$ content ($2.02 \pm 0.12) \times 10^{13}$ atom/kg, which is three orders of magnitude higher than sediment collected from the North Baltic Sea region (Supplementary Table 2). The $233\text{U}/236\text{U}$ atomic ratio ($0.36 \pm 0.05) \times 10^{-2}$ for the Studsvik sediment clearly indicates a higher contribution of reactor input compared to the other five sediments collected in the Baltic Sea with $233\text{U}/236\text{U}$ ratios between $0.59 \times 10^{-2}$ to $0.83 \times 10^{-2}$.

Even though the release of $236\text{U}$ from Studsvik is not well documented due to its low specific radioactivity, it is not surprising that waste discharges from Studsvik contain $236\text{U}$. The high $236\text{U}$ levels in the sediment samples measured most likely originate from scavenging of waterborne $236\text{U}$ from liquid waste discharges by particles into the sediment. Waste dumping/discharge in the Studsvik area are our most plausible candidates for the excess $236\text{U}$ in the Baltic Sea.

Outlooks for future study. The radiological risk associated with $235\text{U}$, $236\text{U}$, and $129\text{I}$ observed in this work is negligible due to their low specific activities and radioactivities. However, the observed unknown $234\text{U}$ reactor source may be an indication of leakage from a previously unrecognized (or unreported) additional radioactive source in the Baltic Sea, e.g., disposed nuclear waste.\textsuperscript{41} Even though the release of $236\text{U}$ from Studsvik is not very well documented (Fig. 1 and Supplementary Table 2) during the COMBINE 2 cruise in 2016. One sediment sample collected outside Studsvik AB in Bergasundet, Bergas strait (58.75 $^\circ$N, 17.40 $^\circ$E) in 2014, which was obtained by pooling 25 sediment slugs (0–10 cm) and homogenized at Swedish Radiation Safety Authority (SSM). The Bergasundet, Bergas strait was the drainage area of the nuclear research facility (Studsvik AB). Details of the sampling campaigns and location of samples are summarized in Supplementary Tables 1 and 2 and Fig. 1.

Standards and reagents. Uranium standard solution (1.000 g/l in 2 M HNO$_3$) purchased from NIST (Gaithersburg, MD) was used after dilution as a standard for the ICP-MS measurement to quantify $238\text{U}$ in seawater. All reagents used in the experiment were of analytical reagent grade and prepared using ultra-pure water (18 M$	ext{cm}$, Milli-Q Advantage A10, Millipore, Bedford, MA) after 10–50 times dilution with 0.5 M HNO$_3$ and 0.1 M NH$_3$·H$_2$O, respectively. The ICP-MS instrument was equipped with an X-series Skimmer cone and a concentric nebulizer under hot plasma conditions. The in-house $238\text{U}$ standards Vienna-KK (238U/235U = (6.89 ± 0.32) × 10$^{-1}$) and Vienna-US (238U/235U = (1.01 ± 0.03) × 10$^{-1}$) diluted by ion (U/Fe = 1:30) were used to monitor the accuracy of the AMS measurement. Five standard samples (3 Vienna-US and 2 Vienna-KK) were measured with a batch of around 30 environmental samples. The Vienna-KK also serve as machine blank for the detection of $233\text{U}$ by AMS.

Analytical methods for determination of $238\text{U}$, $234\text{U}$, $233\text{U}$, $127\text{I}$, and $129\text{I}$. The concentration of $234\text{U}$ and $129\text{I}$ in seawater was measured by ICP-MS (X Series II, Thermo Fisher Scientific, Waltham, MA) after 10–50 times dilution with 2 M HNO$_3$ and 0.1 M NH$_3$·H$_2$O, respectively. The ICP-MS instrument was equipped with an X-series Skimmer cone and a concentric nebulizer under hot plasma conditions. The in-house $238\text{U}$ standards Vienna-KK (238U/235U = (6.89 ± 0.32) × 10$^{-1}$) and Vienna-US (238U/235U = (1.01 ± 0.03) × 10$^{-1}$) diluted by ion (U/Fe = 1:30) were used to monitor the accuracy of the AMS measurement.

The radiochemical method for $233\text{U}$ and $236\text{U}$ separation from seawater was applied according to Qiao et al.\textsuperscript{45} Each seawater sample (0.8–10 l) was filtered with filter paper (Munktell 00 K, particle retention 5–6 μm) to remove large particles and then acidified to pH 2 with concentrated HNO$_3$. Purified FeCl$_3$ solution (0.05 g/ml of Fe) was added to a final Fe concentration of 0.1 g/l. The sample was vigorously stirred with air bubbling for 5–10 min in order to decompose carbonate complexes. In total, 10% NH$_3$·H$_2$O was slowly added to adjust the pH to 8–9 for the co-precipitation of U with Fe(OH)$_3$. The precipitate was evaporated to settle for 0.5–1 h in order to decant most of the supernatant. The sample slurry was centrifuged at 3000 x g for 5 min and the supernatant was discarded. The final residue was dissolved with 15 ml of 3 M HNO$_3$ and the solution was loaded onto a 2 ml UTEVA column which was preconditioned with 20 ml of 3 M HNO$_3$. The UTEVA column was rinsed with 40 ml of 3 M HNO$_3$, followed by 20 ml of 6 M HCl. Uranium absorbed on the column was eluted with 10 ml of 0.025 M HCl. The flow rate for the chromatographic separation was controlled manually to 1.0–1.5 ml/min.

A 100-μl aliquot of U eluate from the column separation was taken for measurement of $235\text{U}$ by ICP-MS to evaluate the chemical yields by comparison with the ideal theoretical analysis on dilute seawater (1–7.9%) from the Baltic Sea (water salinity around 6.0%). The remaining fraction was used to prepare target for AMS measurement of $234\text{U}/238\text{U}$ and $233\text{U}$.\textsuperscript{46} For sediments, 5–10 g of each sample slurry was centrifuged at 3000 x g for 1 h in order to decant most of the supernatant. The sample slurry was centrifuged at 3000 x g for 1 h in order to decant most of the supernatant. The sample slurry was centrifuged at 3000 x g for 1 h in order to decant most of the supernatant. The sample slurry was centrifuged at 3000 x g for 1 h in order to decant most of the supernatant.
A 100-μl aliquot leachate was taken for direct measurement of 238U by ICP-MS, 100 ml of aqua regia on a hotplate for 30 min at 150 °C and then 2 h at 200 °C. A 100-μl aliquot of leachate that was taken for determination of 232Th by ICP-MS, which was used to calculate the 238U concentration in the sediment sample. The remaining leachate was processed following the same procedure (i.e., Fe(OH)3 precipitation and UTEVA column separation) as for seawater samples. The AMS measurement was carried out at the 3-MV tandem accelerator facility Vienna University of Natural Resources (VERA) at the University of Vienna, Austria. To summarize, U, which is extracted as UO2 from a cesium sputter ion source, has to pass a first mass separation stage before it is injected into a tandem accelerator. For the analysis of actinides, the accelerator is operated at a terminal voltage of 2.3 MeV and a ratio of 1.0 × 10−4. All samples, blanks, and standards were measured for six cycles and 5 min per cycle. It should be noted that only the samples collected in 2015 by research vessel Argo were analyzed for 129I. Other samples were not feasible for 129I analysis, since the samples have been acidified before receiving, resulting in loss of iodine due to its high volatility in acidic conditions.

Data availability

The data that support the findings of this study are available on request from the corresponding author up to reasonable request.

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