

A previously unknown source of reactor radionuclides in the Baltic Sea, identified by 233 , 236 , ^{238}U and 127 , ^{129}I multi-fingerprinting

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Abstract

We present the first application of multi-isotopic fingerprints (i.e., $^{236}\text{U}/^{238}\text{U}$, $^{233}\text{U}/^{236}\text{U}$, $^{236}\text{U}/^{129}\text{I}$ and $^{129}\text{I}/^{127}\text{I}$) for the discovery of unrevealed radioactive sources. Our data indicate that, besides the reactor signature from the two European reprocessing plants and global fallout signature, there must be a previously undiscovered additional reactor ^{236}U source in the Baltic Sea. This reactor ^{236}U may come from unreported discharges from nuclear research facilities in Sweden, or it may come from accidental leakage from disposal of spent nuclear fuel on the Baltic seafloor, either reported or unreported. Such leakage would indicate potential problems with the safety of seafloor disposal, and may be accompanied by leakage of other radionuclides. The results demonstrate the high sensitivity of the multi-isotopic tracer systems, especially the newly accessible $^{233}\text{U}/^{236}\text{U}$ signature, to distinguish environmental emissions of unrevealed historical or present radioactive releases for nuclear safeguard and emergency preparedness, as well as tracing environmental processes from the releasing sites.

Introduction

^{236}U ($t_{1/2} = 2.34 \times 10^7$ y) is an isotope of uranium, which is produced by thermal neutron capture of the omnipresent ^{235}U via (n, γ)-reactions and through ^{238}U (n, 3n) ^{236}U reactions with fast neutrons. Even though a small amount (about 35 kg in total) of ^{236}U is produced naturally in the Earth's surface environments, ^{236}U is (by mass) the largest secondary product created in nuclear reactors, estimated totally to be an order of 10^6 kg¹. ^{236}U is a sensitive tracer of deliberate or accidental leakage from the nuclear fuel/waste cycle²⁻⁵. The known sources of reactor ^{236}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 currents⁶. Emissions from other known sources of reactor ^{236}U , e.g., the Springfield nuclear facility and the Fukushima accident, are negligible^{5,7}.

A significant amount of ^{236}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 1960s⁸. This omnipresent fallout source can make identification of unreported sources of reactor ^{236}U challenging, because of methodological difficulties in distinguishing the source of ^{236}U ⁹. In addition, the $^{236}\text{U}/^{238}\text{U}$ ratio does not provide source information because of ^{238}U of natural origin is ubiquitous.

Reactor ^{236}U could be differentiated from fallout ^{236}U because these sources have different and characteristic $^{233}\text{U}/^{236}\text{U}$ ratios due to different nuclear production mechanisms. ^{233}U was mostly produced during nuclear weapons testing by fast neutrons via ^{235}U (n, 3n) ^{233}U reactions or directly by ^{233}U -fueled devices, whereas almost no ^{233}U is produced in thermal nuclear power reactors or reprocessing plants¹⁰. Recently ^{233}U measurements at environmental level have become possible¹⁰.

The representative $^{233}\text{U}/^{236}\text{U}$ atomic ratio of global fallout from atmospheric nuclear weapons testing was suggested to be $(1.40 \pm 0.12) \times 10^{-2}$ ¹⁰. This is several orders of magnitude higher than the $^{233}\text{U}/^{236}\text{U}$ atomic ratio in nuclear reactors, e.g., 1×10^{-7} - 1×10^{-6} in LH discharges¹¹, which agrees well with reactor model calculations¹². 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 measured⁹, reflecting a dominant reactor signal released from SF. The use of the $^{233}\text{U}/^{236}\text{U}$ atomic ratio helps better distinguishing the origin of ^{236}U and since being radionuclides of the same element, the $^{233}\text{U}/^{236}\text{U}$ ratio will not be affected during the transport pathway. In addition, the combination of ^{236}U with other radionuclides, e.g. ^{129}I , can be useful to trace the transport of ^{236}U from specific source points, e.g., releases from LH and SF¹³⁻¹⁶.

The Baltic Sea is a highly polluted sea, including potentially for anthropogenic radionuclides. It receives radionuclides from global fallout, discharges from the two European reprocessing plants, potentially from the Chernobyl accident, and from any other local sources. In this study, we use a novel combination of three anthropogenic radionuclides – ^{233}U , ^{236}U , and ^{129}I - to identify a previously unknown local source of radionuclide pollution to the Baltic Sea.

Results

The study area and sampling. The Baltic Sea is a landlocked intracontinental sea in Northern Europe and constitutes one of the largest brackish water environments on Earth with about 80 million inhabitants in the surrounding states¹⁷. 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 (Figure 1). The driving force for the water circulation is fresh water surplus from river run-off estimated at $473 \text{ km}^3 \text{ yr}^{-1}$ together with “recycled” North Sea inflowing water as Baltic outflow that sum up to a total water exchange rate of $753 \text{ km}^3 \text{ yr}^{-1}$ ¹⁸. A mean residence time for the 21580 km^3 Baltic water volume was estimated to be 29 years which is equivalent to a “half-life” for the water volume of 20 years¹⁸.

In the investigation presented here, water and sediments samples were collected from the Baltic Sea and related water masses including the western Danish coast in the years 2011-2016 (Table S1 and S2). The water sampling covers mainly the surface distribution (0-5 m depth), with a few samples from deep water and one riverine water from the Mälaren river, which receives downstream discharges from a nuclear fuel fabrication facility (Westinghouse) in Sweden. In addition to the Baltic Sea water, we analyzed sediment samples to gain some idea about the accumulation trend of the isotopes in the bottom of 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 (Figure. 1) in the Baltic Sea including 1) KGR: Kattegat-Skagerrak and the region including the 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, Borholm 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.

Distribution of ^{236}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 (Table S1, S2) vary within $(5-52) \times 10^{-9}$, with the higher ratios (in the central and northern parts of the Baltic Sea and lower ones in the western parts (Danish Straits, Kattegat/Skagerrak and Danish west coast). The highest value reported here is 6-fold higher than the average value found in the North Sea in 2010 $((7.6 \pm 3.7) \times 10^{-9})$ ¹⁹.

The distribution patterns (Figure 2) suggest a decline of ^{236}U concentration that is labeled with discharges from LH and SF in the North Sea at the water crossover into the Kattegat and further into the Baltic Sea. However, high ^{236}U concentrations $((6-9) \times 10^7 \text{ atom/L})$ are observed in the surface water of the Bothnian Sea and Bothnian Bay, which are comparable to the central North Sea values $((3-10) \times 10^7 \text{ atom/L})$ ¹⁹. 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 increasing pattern of $^{236}\text{U}/^{238}\text{U}$ ratio points out an additional, likely local, supply of ^{236}U in the Baltic Sea ⁷.

$^{233}\text{U}/^{236}\text{U}$ atomic ratios obtained here are in the range of $(0.14-0.87) \times 10^{-2}$, with the lower $^{233}\text{U}/^{236}\text{U}$ atomic ratios distributed close to the western parts of the Baltic, including the Danish coast, and the higher ratios in the central Baltic Sea. As the typical $^{233}\text{U}/^{236}\text{U}$ ratio for global fallout is $(1.4 \pm 0.1) \times 10^{-2}$ ⁹, 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}I concentration, $^{129}\text{I}/^{127}\text{I}$ and $^{236}\text{U}/^{129}\text{I}$ atomic ratios

The measured ^{129}I concentrations $((3-232) \times 10^9 \text{ atom/L})$ 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 ²⁰, with the highest values in the North Sea-Skagerrak-Kattegat which decrease toward the Sound and remain relatively constant in the Baltic Proper. The distributions of ^{129}I concentrations and $^{129}\text{I}/^{127}\text{I}$ atomic ratios indicate that the major source of ^{129}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 ²¹.

Aldahan et al. ²¹ reported that the average concentration of ^{129}I in the rivers around the Baltic Sea was $3.9 \times 10^8 \text{ atom/L}$, which suggested some minor contribution of ^{129}I from riverine water to the Baltic Sea. The ^{129}I concentrations show a larger gradient (two orders of magnitude) compared to the ^{236}U concentrations (15-fold) along the Baltic Sea. $^{236}\text{U}/^{129}\text{I}$ ratios are within the range of $(5-133) \times 10^{-4}$ and

indicate a reversed geographical distribution compared to ^{129}I concentration and $^{129}\text{I}/^{127}\text{I}$ atomic ratio (Figure 2).

Discussion

Potential sources of uranium and iodine in the Baltic Sea

The overall sources of uranium and iodine in the Baltic Sea can be summarized as follows:

- A) Natural ocean water, with salinity of 35‰, which contains about 60 µg/L of ^{127}I , 3 µg/L of ^{238}U , but negligible ^{129}I , ^{236}U and ^{233}U .
- B) Natural fresh water with salinity < 1‰ and negligible concentrations of ^{129}I , ^{236}U and ^{233}U , and lower ^{127}I and ^{238}U than seawater (0.05-10 µg/L for both nuclides).
- C) Global fallout from atmospheric nuclear weapons testing, with negligible ^{127}I and ^{238}U , an average $^{233}\text{U}/^{236}\text{U}$ atomic ratio of $(1.4 \pm 0.2) \times 10^{-2}$, and a surface geographical distribution pattern for ^{236}U and ^{233}U similar to that of ^{137}Cs which is not unusually high in the Baltic region²². Earlier studies have estimated ^{236}U concentration (up to 1.4×10^8 atom/L peaking in 1960s) in surface water of the North Sea to be related to global fallout, which may have been partly masked by discharges from the nuclear reprocessing of LH and SF^{21,22}. In the Baltic Sea, that has 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 $^{233}\text{U}/^{236}\text{U}$ 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 $^{233}\text{U}/^{236}\text{U}$ atomic ratio stays constant.
- D) Marine discharges from European nuclear fuel reprocessing plants (including mainly SF and LH), with known ^{236}U and ^{129}I source functions^{23,24}, 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 ^{233}U should also be absent from marine discharges of the reprocessing plants.
- E) The Chernobyl accident. Pu from Chernobyl has been found in fallout over central Europe²⁵ 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 $^{236}\text{U}/^{238}\text{U}$ and $^{233}\text{U}/^{236}\text{U}$ atomic ratios set by the balance of reprocessing discharge and global fallout^{9,19}. As they distribute in the Baltic and mix with waters from various rivers, these 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}U source identification via binary mixing

The concentration of ^{238}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}U concentration of $0.33 \pm 0.05 \mu\text{g/L}$, which falls in the range ($0.2\text{--}0.7 \mu\text{g/L}$) of ^{238}U for some rivers in the Baltic Sea region²⁶. There is more scatter in the ^{238}U concentration for low salinities, which might be attributed to differences in regional riverine input. I-129 also shows a positive linear correlation ($R^2 = 0.69$) with salinity (Fig. 3B), but strong scatter occurs at the high salinity end. This trend can be attributed to the mixing of ^{129}I enriched North Sea coast water with ^{129}I depleted North Atlantic water in the Kattegat-Skagerrak region (Fig. 1 A). The ^{238}U and ^{129}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 river-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 water 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 from an average of $(8 \pm 2) \times 10^{-4}$ in the Kattegat-Skagerrak region, corresponding to reprocessing derived ^{236}U and ^{129}I , to 1×10^{-2} in the central Baltic Sea. Both ratios indicate addition of ^{236}U from a local source. The difference in increases between the two ratios can be explained by the presence of ^{238}U and possibly ^{129}I in that local source in addition to ^{236}U . If the source does not contain any ^{129}I , the 10-fold increase in $^{236}\text{U}/^{129}\text{I}$ suggests that ca. 90% of ^{236}U in the central Baltic Sea is from local sources. If the source does contain ^{129}I , the portion of ^{236}U derived locally must be still larger.

To understand the source terms of ^{236}U in the Baltic Sea, a binary mixing model is applied with two respective end members representing ^{236}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 (Table S3)^{19,27}. The deviation of the observed $^{236}\text{U}/^{238}\text{U}$ atomic ratio from the binary mixing (line L1, Fig. 4A) between the North Sea water and an assumed freshwater end member containing no ^{236}U (neither ^{233}U) reflects additional ^{236}U sources besides North Sea water. The spatial distribution of deviations in the $^{236}\text{U}/^{238}\text{U}$ atomic ratio allow locating the additional ^{236}U source (Figure S2). The distribution pattern in Figure S2 is compatible with the hypothesis of additional riverine ^{236}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 some global fallout may still be washing from the land surface. The $^{236}\text{U}/^{238}\text{U}$ and $^{236}\text{U}/^{129}\text{I}$ ratios therefore do not directly indicate whether the excess ^{236}U is only from global fallout, or from an additional, previously undiscovered, source that has directly released ^{236}U to the Baltic Sea.

Application of $^{233}\text{U}/^{236}\text{U}$ atomic ratio for ^{236}U source identification

If we assume that the excess ^{236}U originates only from global fallout, the $^{236}\text{U}/^{238}\text{U}$ atomic ratio of the riverine input in the best-fit binary mixing is 6×10^{-8} (line L, Fig. 4A). However, there is a clear deviation of the observation from the model for $^{233}\text{U}/^{236}\text{U}$ atomic ratios (Fig. 5A). A subgroup of samples from the Kattegat-Skagerrak reveal a relatively stable $^{233}\text{U}/^{236}\text{U}$ atomic ratio of 0.2×10^{-2} (blue dash-dotted line in Fig. 5) independent of $^{236}\text{U}/^{238}\text{U}$ and salinity. This behavior can be explained by assuming an end member of North Sea water with $^{233}\text{U}/^{236}\text{U}$ atomic ratio = 0.2×10^{-2} (a mixed signal of global fallout plus nuclear reprocessing) and salinity 35‰, 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 middle and north Baltic Sea region indicate a typical $^{233}\text{U}/^{236}\text{U}$ atomic ratio of 0.5×10^{-2} (the green dash-dotted line in Fig. 5), uncorrelated with both $^{236}\text{U}/^{238}\text{U}$ and salinity. This cluster lies significantly below the binary mixing model L, indicating an additional local ^{236}U sources besides the global fallout, which is characterized by low $^{233}\text{U}/^{236}\text{U}$ atomic ratio. A low $^{233}\text{U}/^{236}\text{U}$ 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-third of the anthropogenic uranium observed in the middle and north Baltic Sea region seems to originate from this third source (Equation (1)), indicating a strong contribution of ^{236}U without ^{233}U , e.g. a nuclear reactor sourced ^{236}U .

$$R_s = \frac{N_{233,f} + N_{233,r}}{N_{236,f} + N_{236,r}} = \frac{N_{236,f} \cdot R_f + N_{236,r} \cdot R_r}{N_{236,f} + N_{236,r}} = \frac{R_f + \frac{N_{236,r}}{N_{236,f}} \cdot R_r}{1 + \frac{N_{236,r}}{N_{236,f}}} \quad (1)$$

Where R_s , R_f and R_r represent respectively the $^{233}\text{U}/^{236}\text{U}$ atomic ratio of the Baltic seawater, global fallout and nuclear reactor; $N_{233,f}$ and $N_{233,r}$ refer to the atomic number of ^{233}U from global fallout and reactor, respectively; $N_{236,f}$ and $N_{236,r}$ refer to the atomic number of ^{236}U from global fallout and reactor, respectively. Therefore, $\frac{N_{236,r}}{N_{236,f}} = \frac{R_f - R_s}{R_s - R_r}$. With $R_s = 0.5 \times 10^{-2}$, $R_f = 1.4 \times 10^{-2}$ and $R_r = 0.12 \times 10^{-2}$, we obtain that the ^{236}U contribution from our assumed reactor source is 2.4 times that of global fallout.

To locate this additional reactor ^{236}U source, we apply another binary mixing line L2 (Figure 4 A) of the North Sea water with riverine water, the latter carrying global fallout which accounts for $1/(1+2.4)$ of the average ^{236}U concentration in the Baltic Sea ($(6.0 \pm 1.7) \times 10^7$ atom/L). Thus, the riverine endmember is characterized by salinity =0, $^{238}\text{U} = 0.4 \mu\text{g/L}$, $^{236}\text{U} = 1/(1+2.4) \times (6 \times 10^7) = 2 \times 10^7$ atom/L, and $^{236}\text{U}/^{238}\text{U}$

atomic ratio = 2×10^{-8} . The excesses of $^{236}\text{U}/^{238}\text{U}$ atomic ratio from the mixing curve L2 and their spatial distribution are demonstrated in Figure 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 ^{236}U unknown source

To identify the source of the excess ^{236}U , the order of magnitude of ^{236}U inventories and fluxes must be estimated. It should be noted this calculation is based only on our data on surface waters, and a precise interpretation will require substantially more data, and to account for many different effects such as vertical distribution of ^{236}U in the Baltic water columns and on the scavenging of uranium into the sediment (especially in the anoxic regions).

The median salinity of the Baltic Sea seawater analyzed here is about 8.3 ‰ (comparable to the reported value of 8.6 ‰ for the Baltic outflow water¹⁸), meaning that the ratio of seawater to riverine water is 1:4. An average excess of 6×10^7 atom/L ^{236}U in riverine water is obtained based on the deviation of the ^{236}U concentration from L1 in samples from NBR (Figure S2). The volume of the Baltic Sea of 21700 km³ with 80% riverine water corresponds to 400 g of ^{236}U . Taking into account that ca. 71% (i.e., $N_{236,r}/N_{236,f}=2.4$) of this excess ^{236}U is from the additional reactor source (~280 g) as discussed above, the remainder (~120 g) is related to global fallout.

It is estimated that a total inventory of 1000 kg of anthropogenic ^{236}U was distributed via global fallout during the 1950s and 1980s mainly on the Northern Hemisphere⁷. Considering the surface area of the Baltic Sea of 3.77×10^5 km² (without the catchment area) in comparison to the Northern Hemisphere (half of the Earth's surface area, i.e. 5.10×10^8 km²), then the total ^{236}U deposition from direct global fallout is estimated as 1.5 kg.

However, when considering the 29-year mean residence time of Baltic seawater, then most of the 1.5 kg ^{236}U was transported out after 60 years, leaving behind 0.19 kg. In addition, some particle-associated ^{236}U fraction from global fallout might be incorporated into the Baltic sediment²⁹. Therefore, the above estimation of 120 g remaining ^{236}U in the Baltic seawater from global fallout (using salinity data) seems justified.

Emissions from the Chernobyl accident may be an additional ^{236}U source in the Baltic Sea, yet it is difficult to be identified. 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 limited documentation about the ^{233}U and ^{236}U release records from these installations (Table S4)¹¹. Data on ^{236}U is available from Westinghouse during 1998–2017, with a total reported release of 1.06×10^6 Bq of ^{236}U , equal to 0.44 g. In addition, we measured two seawater samples collected in Mälaren River (Table S2), which receives waste discharges from the Westinghouse facility.

The results show that the $^{236}\text{U}/^{238}\text{U}$ ratios is at the level of 2×10^{-8} , which is comparable with the seawater samples collected in the central Baltic Sea. The river 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 0.44 g of ^{236}U released from the Westinghouse installation is negligible compared to the above estimated 280 g of the unknown reactor source in the Baltic Sea. For the Mälaren river, the ^{238}U concentration was measured to be $1.5 \pm 0.1 \mu\text{g/L}$ in this work, together with a flux of $166 \text{ m}^3/\text{s}$ ²⁸, it means an input of 0.1 g/yr of ^{236}U , which is negligible also.

Another candidate we assume is reactor fuel, dumped into the sea; the atomic ratio of $^{236}\text{U}/^{238}\text{U}$ can be as high as 1×10^{-2} in conventional nuclear reactors, which would require only 27 kg of dumped/dissolved fuel (a commercial nuclear reactor contains ~ 100000 kg of fuel). ^{235}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 reactors³⁰. The concentration will be even higher in the core of a nuclear reactor for marine applications, where enriched or highly enriched ^{235}U is used; the Russian submarine cores reportedly contain some 50 to 200 kg of ^{235}U ³¹. The former Soviet Union (USSR) was accused for dumping radioactive waste in the Baltic Sea, yet it is not possible to assess the dumped amount^{32,33}.

The geographical distribution of $^{236}\text{U}/^{238}\text{U}$ atomic ratio in surface seawater of central Baltic Sea shows high values nearby the Swedish coast close to Stockholm, where a nuclear research company Studsvik AB, Nyköping (100 km south of Stockholm), Sweden, 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 Studsvik³⁴. Our measurement on some sediment samples from the Studsvik area show very high ^{236}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 (Table S2). 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 0.59×10^{-2} to 0.83×10^{-2} .

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

Methods

Detailed description of the study area and sampling. The Baltic Sea features three major basins, the Bothnian Bay, the Bothnian Sea and the Baltic Proper. The two northerly basins (Bothnian Bay and

Bothnian Sea) are characterized by low salinity water mass (1-3 ‰ and 3-7 ‰, respectively) and rather weak vertical salinity stratifications, although strong thermoclines usually develop during the summer³⁵. The Bothnian Sea represents a large reservoir of brackish water mass that can be divided into two layers blocked by a weak halocline around a depth of 60 m. The long term circulation of the Bothnian Sea water is dominated by an estuary circulation where the bottom dense waters can be traced as surface water in the Baltic Proper³⁶. The Baltic Proper is the largest basin in the Baltic Sea, permanently stratified in the central part with a strong halocline around a depth of 75 m separating the surface water (salinity 7-8 ‰) from the deep water (salinity 9-20 ‰) and a long-term cyclonic current circulation pattern³⁷. Water exchange in the Baltic Proper happens through renewing of the deep water during extreme inflow events from the Kattegat. The water mass circulation is further associated with outflow of surface water to the Kattegat and inflow of fresher surface waters from the Bothnian Sea, the Gulf of Finland and the Gulf of Riga (Figure 1).

Water samples analysed in the present investigation were collected via different cruises during 2011-2016. Samples of 2011 were obtained from GEOTRACES cruise on board of research vessel R/V Oceania. Samples from 2013-2014 were collected through the environmental monitoring program for Helsinki Commission (HELCOM). Samples from 2015 were collected on board the research vessel Argos, operated by the marine division of the Swedish Metrological and Hydrological Institute (SMHI). Samples from 2016 were obtained from the Radiation and Nuclear Safety Authority (STUK), Finland, through sampling cruise COMBINE 2 on the research vessel R/V Aranda. One riverine water sample from Mälaren river (in Sweden: 59.33 °N, 18.04 °E) was also sampled for the radioisotope analyses, as this river receives downstream discharges from a nuclear fuel fabrication facility (Westinghouse) in Sweden.

Five surface (0-2 cm) sediments in the middle and north parts of Baltic region were collected (Figure 1 and Table S2) during the COMBINE 2 cruise in 2016. One sediment sample collected outside Studsvik in Bergasundet, Bergas strait (58.75 °N, 17.40 °E) in 2014, which was obtained by pooling 25 sediment plugs (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). Details of the sampling campaigns and location of samples are summarized in Table S1, S2 and Figure 1.

Standards and reagents. Uranium standard solution (1.000 g/L in 2 mol/L HNO₃) was purchased from NIST (Gaithersburg, MD), which was used after dilution as a standard for the ICP-MS measurement to quantify ²³⁸U in seawater. All reagents used in the experiment were of analytical reagent grade and prepared using ultra-pure water (18 MΩ·cm). UTEVA resin (100-150 μm particle size) was purchased from Triskem International, Bruz, France and packed in 2-mL Econo-Columns (0.7 cm i.d. × 5 cm length, Bio-Rad Laboratories Inc., Hercules, CA) for the chemical purification of uranium isotopes.

Analytical methods for determination of ²³⁸U, ²³⁶U, ²³³U, ¹²⁷I and ¹²⁹I. The concentration of ²³⁸U and ¹²⁷I in seawater was measured by ICP-MS (X Series^{II}, Thermo Fisher Scientific, Waltham, MA) after 10-50 times dilutions with 0.5 M HNO₃ and 0.1 M NH₃·H₂O, respectively. The ICP-MS instrument was equipped with an Xt-skimmer core and a concentric nebuliser under hot plasma conditions. The typical operational

conditions of the instrument have been given elsewhere³⁸. Indium (as InCl_3) was used as an internal standard and 0.5 mol/L HNO_3 solution was used as a washing solution between consecutive assays.

The radiochemical method for ^{233}U and ^{236}U determination in seawater was applied according to Qiao et al.³⁹. In short, uranium in each seawater sample (0.8-10 L) was co-precipitated with $\text{Fe}(\text{OH})_3$, followed by purification with a 2-mL UTEVA column. A 100- μL aliquot of U eluate from the column separation was taken for measurement of ^{238}U by ICP-MS to evaluate the analytical chemical yields by dividing the values in the original samples. The remaining fraction was prepared as target for the AMS measurement of $^{236}\text{U}/^{238}\text{U}$ and $^{233}\text{U}/^{236}\text{U}$. For sediments, 5-10 g of each dried sample was ashed overnight at 450 °C in a muffle oven and leached with 100 mL of *aqua regia* on a hotplate for 30 minutes at 150 °C and then 2 hours at 200 °C. A 100- μL aliquot was taken from the leachate for directly measurement of ^{238}U by ICP-MS, which was used to calculate the original ^{238}U concentrations in the sediment sample. The remaining leachate was processed following the same procedure (i.e., $\text{Fe}(\text{OH})_3$ co-precipitation and UTEVA column separation) as for seawater samples. The AMS measurement was carried out at the 3-MV tandem accelerator facility VERA (Vienna Environmental Research Accelerator) at the University of Vienna, Austria. The detailed procedure for the sample preparation and AMS measurement of ^{233}U and ^{236}U has been reported elsewhere⁴⁰.

For the determination of ^{129}I in seawater, 100 ml of sample was transferred a separation funnels. 2.0 mg of ^{127}I carrier (prepared using iodine crystal purchased from Woodward company, USA, with a $^{129}\text{I}/^{127}\text{I}$ ratio of 2×10^{-14}), 500 Bq of ^{125}I tracer and 0.5 mL of 0.5 mol/L $\text{Na}_2\text{S}_2\text{O}_5$ solution were added to the funnel, and then the pH of the solution was adjusted to 1-2 using 3 mol/L HNO_3 to convert all iodine species to iodide. With addition of 20-50 mL chloroform (CHCl_3) and 2-5 mL 1.0 mol/L NaNO_2 , iodide was oxidized to I_2 and extracted to CHCl_3 phase by shaking. The extraction procedure was repeated three times to extract all iodine. The CHCl_3 phases were combined to a new funnel, 20 mL H_2O and 0.3-0.5 mL 0.05 mol/L $\text{Na}_2\text{S}_2\text{O}_5$ solution was added to the funnel to reduce I_2 in chloroform phase to iodide and back-extracted iodine into water phase. This extraction and back extraction were repeated once for further purification.

The separated iodine (in iodide form) in a small volume (5-7 mL) was transferred to a centrifuge tube, 1.0 mL of 0.5 mol/L AgNO_3 solution and 1 mL of 3.0 mol/L HNO_3 were added to form AgI precipitate. The AgI precipitate was separated using centrifugation at 3500 rpm for 3-5 min, and washed in sequence using 10 mL 3 mol/L HNO_3 and two aliquots of 10 mL deionized water to remove possibly formed Ag_2SO_3 and Ag_2SO_4 which are soluble in acidic solution. The precipitate was transferred to a 1.5 mL centrifuge tube. ^{125}I in the precipitate was measured using a NaI gamma detector for calculating the chemical yield of iodine. The prepared AgI precipitate in small tube was dried at 70°C and weighed, The dried precipitate was ground to fine powder and mixed with five times by mass of niobium powder (325 mesh, Alfa Aesar, Ward Hill, MA), which was finally pressed into a copper holder using a pneumatic press (Zhenjiang Aode Presser Instruments Ltd.). $^{129}\text{I}/^{127}\text{I}$ atomic ratios in the prepared targets were measured

by the 5 MV AMS system at the Tandem Laboratory, Uppsala University. The standard used in the measurement was the NIST-SRM-4949c ^{129}I . All samples, blanks and standards were measured for 6 cycles and 5 minutes per sample in each cycle. A detailed description of AMS system and measurement of ^{129}I has been reported elsewhere ⁴¹. It should be noted that only the samples collected in 2015 by research vessel Argos were analysed for ^{129}I . Other samples were not feasible for ^{129}I analysis, since the samples have been acidified before receiving, resulting in loss of iodine due to its high volatility in acidic conditions.

Declarations

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Author contributions

J. Q. initiated and coordinated the study, wrote manuscript, performed chemical analysis for uranium and made data evaluation. H. Z. performed chemical analysis for iodine. K. H and P. S performed AMS measurement for uranium. A.A. and G. P. coordinated AMS measurement for iodine. H. Z., G. H., X. H., V. V., A.A. and M. E. contributed to sample collection. G. H. and P. S. have valuable input to the outline of the discussion. All contributed to interpretation and manuscript reviewing.

Competing interests

The authors declare no competing interests.

Additional information

Supporting information includes Tables S1-S4 and Figure S1, which is available online.

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Figures

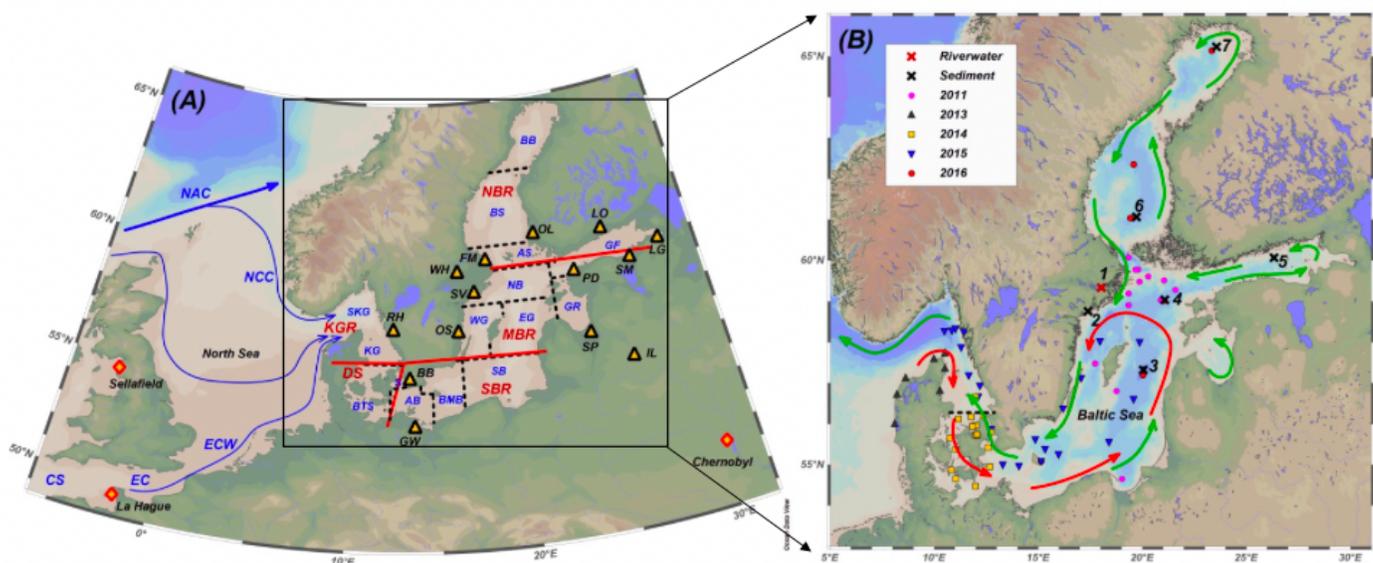


Figure 1

Overview of schematic circulation water mass in North Sea-Baltic Sea region (A) and sampling stations in this work as well as nuclear installations around the Baltic Sea (B). The symbols in (A) are CS: Celtic Sea; EC: English Channel; ECW: English Channel Waters; NAC: North Atlantic Current; NCC: Norwegian Coastal Current; BB: Bothnian Bay; BS: Bothnian Sea; AS: Archipelago and Åland Sea; GF: Gulf of Finland; NB: Northern Baltic Proper; WG: Western Gotland Basin; EG: Eastern Gotland Basin; GR: Gulf of Riga; SB: South Baltic Proper; BMB: Bornholm Basin; AB: Arkona Basin; S: The Sound; BTS: Belt Sea; KG: Kattegat; SKG: Skagerrak; KGR: Kattegat-Skagerrak region including the Jutland west coast nearby the North Sea; DS: Danish Straits including the Belt Seas and the Sound; SBR: South Baltic Sea region including Arkona Basin, Bornholm Basin and South Baltic Proper; MBR: Middle Baltic Sea region including Northern Baltic Proper, Western Gotland Basin, Eastern Gotland Basin and Gulf of Riga; and NBR: North Baltic Sea region including Archipelago and Åland Sea, Bothnian Sea and Bothnian Bay) Nuclear installations including: RH: Ringhals NPP; BB: Barseback NPP; GW: Greifswald NPP; OS: Oskarshamn NPP; SV: Studsvik AB site; WH: Westinghouse Electric Sweden AB; FM: Forsmark NPP; OL: Olkiluoto NPP; LO: Loviisa NPP; LG:

Leningrad NPP; IL: Ignalina NPP; SM: Sillamäe site; PD: Paldiski site; SP: Salaspils research reactor; The stations marked with cross in (B) are either riverwater or sediment samples (1-Marelene river water, 2-Studsvik sediment, 3-sediment BY15; 4-sediment LL17; 5-sediment LL3a; 6-sediment EB1; 7-sediment CVI), all the other samples are seawaters collected in different years during 2011-2016 as marked with different symbols. Red arrows refer to bottom water movement and green arrows refer to surface water movement.

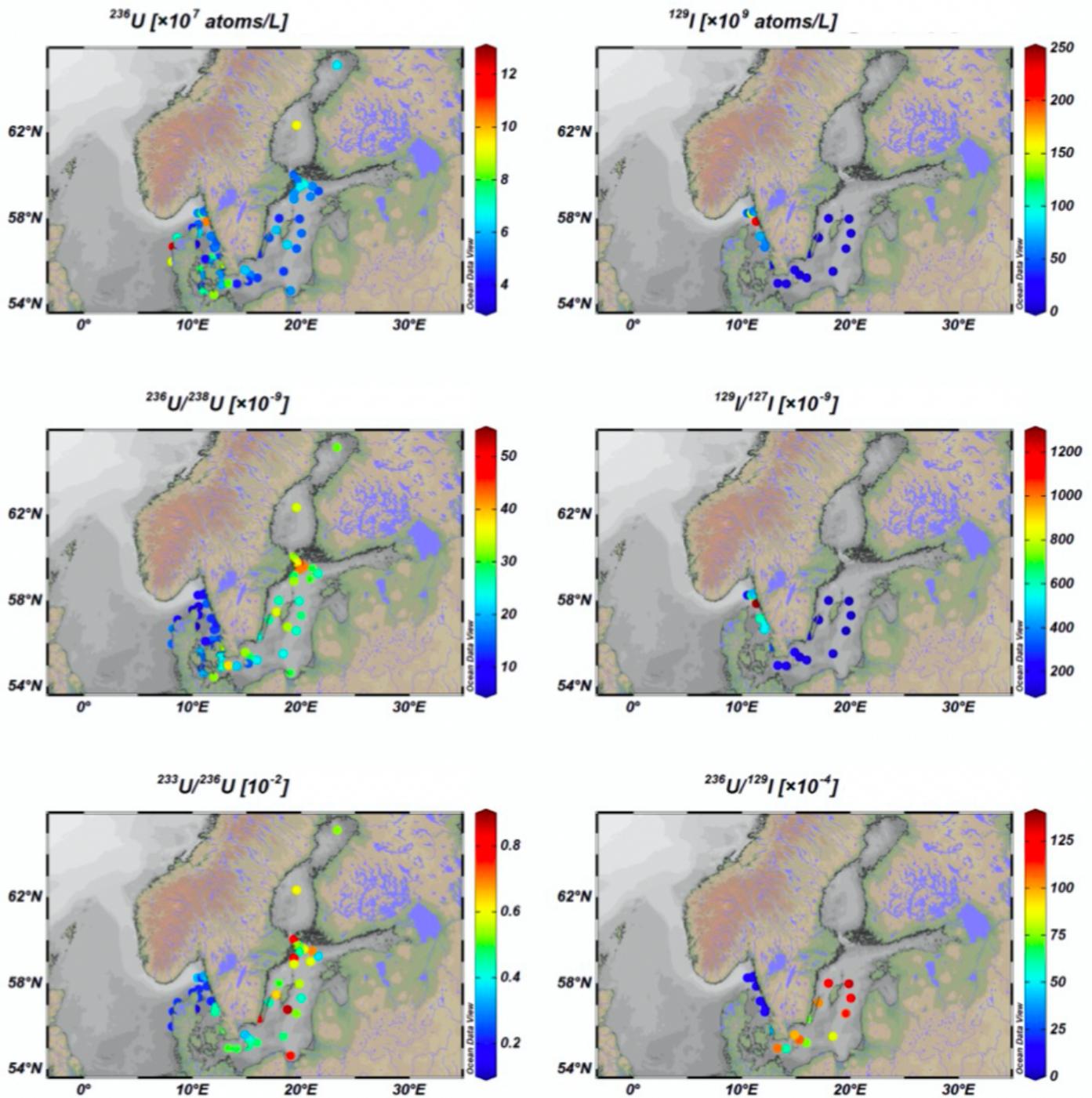


Figure 2

Distribution of ^{236}U and ^{129}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.

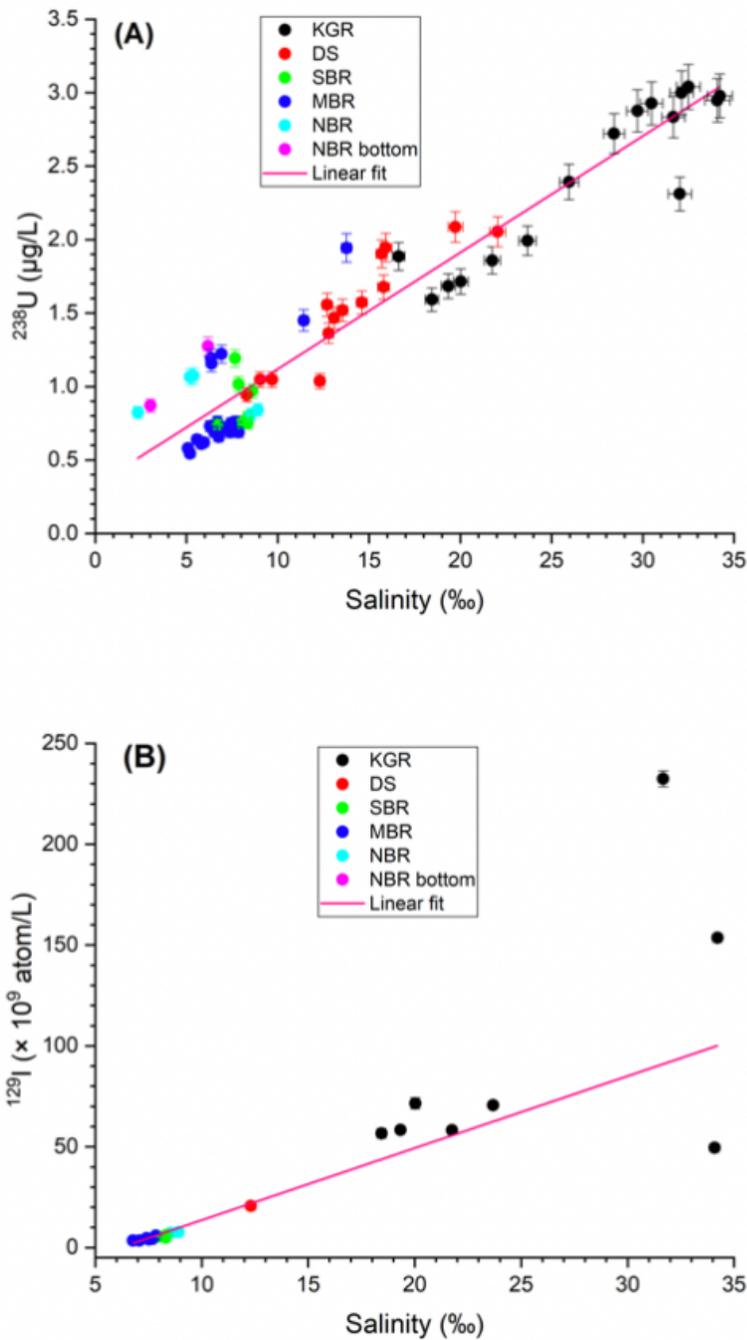


Figure 3

Correlation ^{238}U (A) and ^{129}I (B) concentrations with salinity. (KGR: Kattegat-Skagerrak region including the Jutland 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; and NBR: North Baltic Sea region including Archipelago and Åland Sea, Bothnian Sea and Bothnian Bay).

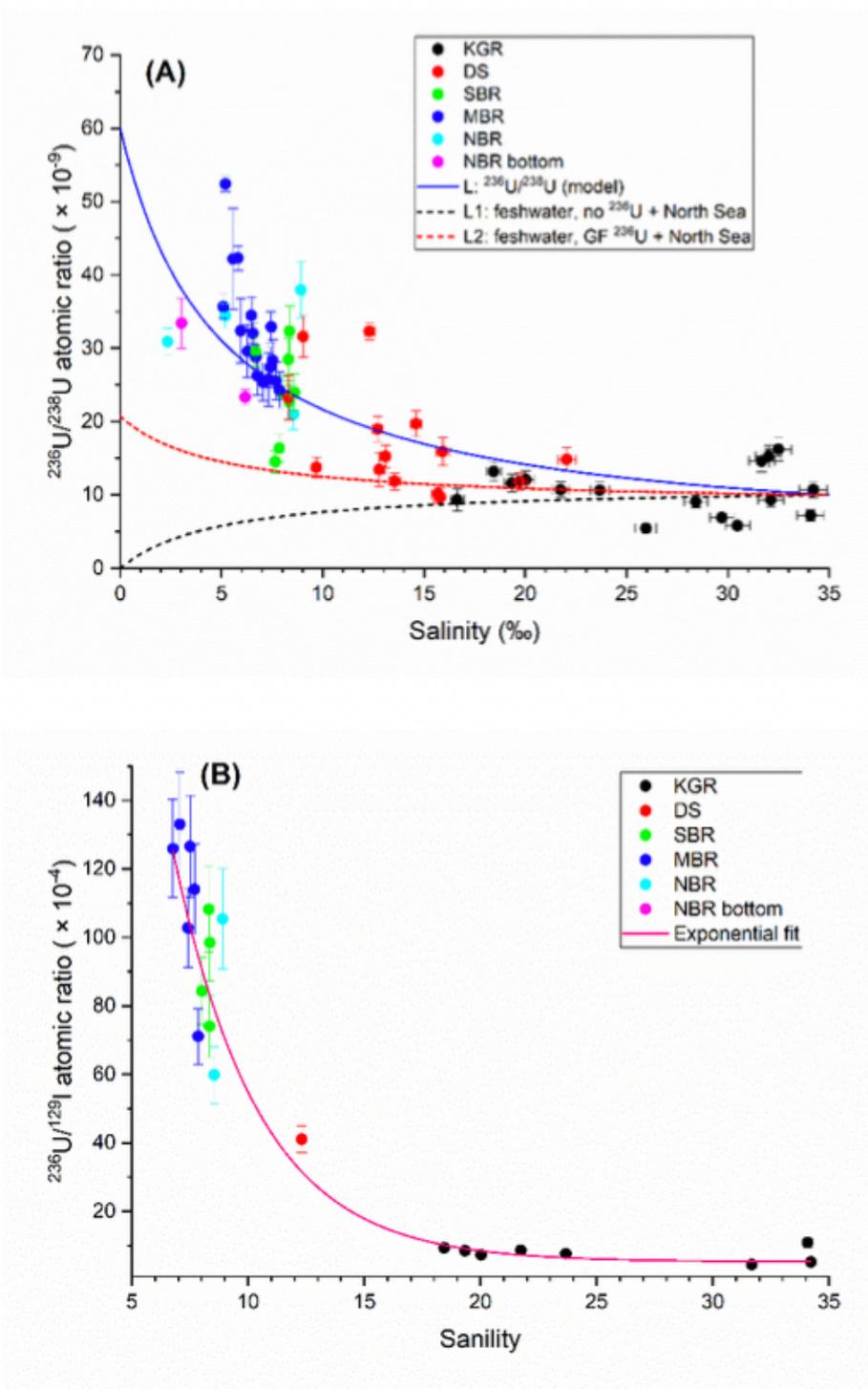


Figure 4

Variation of $^{236}\text{U}/^{238}\text{U}$ atomic ratio (A) and $^{236}\text{U}/^{129}\text{I}$ ratios (B) with salinity. (KGR: Kattegat-Skagerrak region including the Jutland 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; and 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, $^{238}\text{U} = 0.4 \mu\text{g/l}$, $^{236}\text{U}/^{238}\text{U}$ atomic ratio = 6×10^{-8} and $^{236}\text{U} = 6 \times 10^7 \text{ atom/L}$; L1 (black dashed line): the binary mixing line between the North Sea water and an assumed freshwater end member containing no ^{236}U (salinity=0, $^{238}\text{U} = 0.4 \mu\text{g/l}$, $^{236}\text{U}/^{238}\text{U}$ atomic ratio = 0 and $^{236}\text{U} = 0$); L2 (red dashed line): the binary mixing line between the North Sea water and an assumed freshwater end member with salinity =0, $^{238}\text{U} = 0.4 \mu\text{g /L}$, $^{236}\text{U} = 2 \times 10^7 \text{ atom/L}$, and $^{236}\text{U}/^{238}\text{U}$ atomic ratio = 2×10^{-8} ; the pink solid line: exponential fit of $^{236}\text{U}/^{129}\text{I}$ atomic ratio vs. salinity)

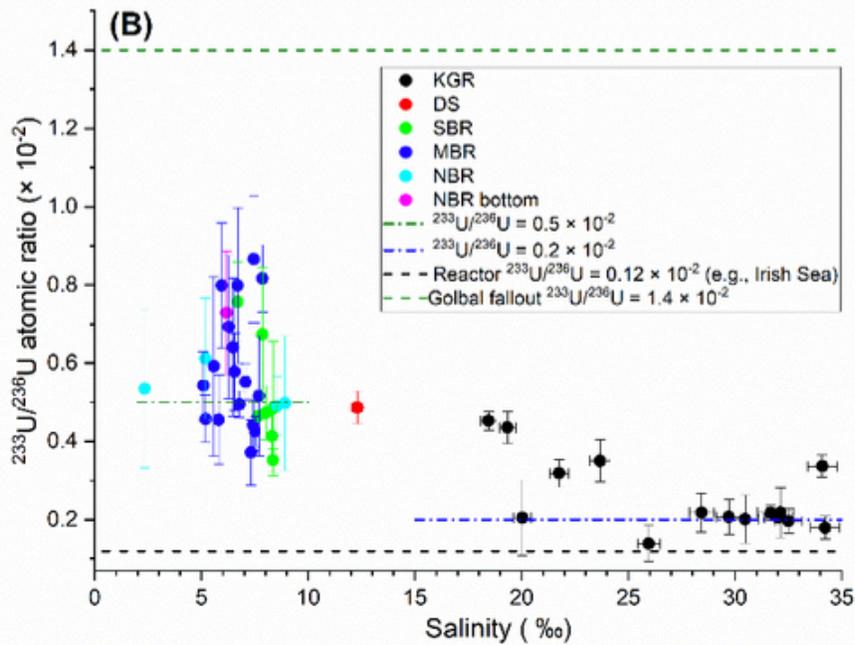
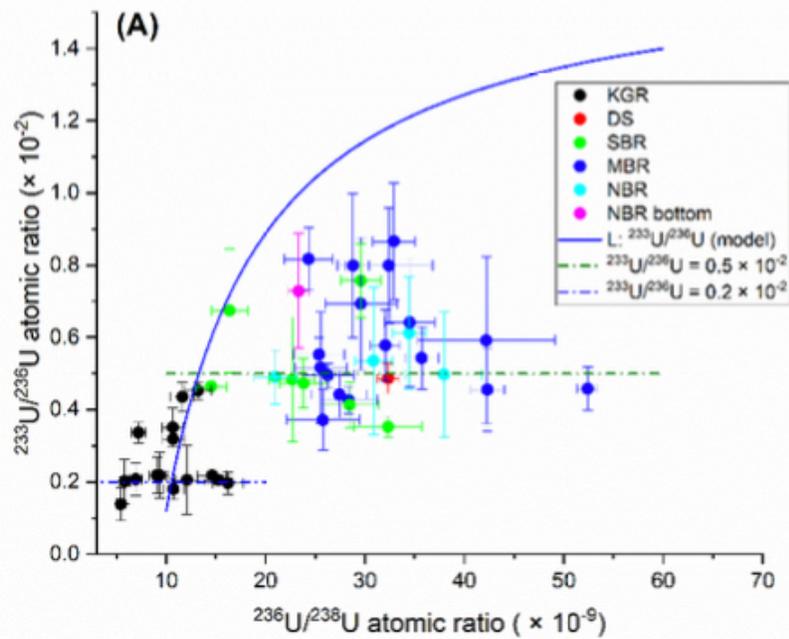


Figure 5

Variations of $^{233}\text{U}/^{236}\text{U}$ atomic ratio with salinity. (KGR: Kattegat-Skagerrak region including the Jutland 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; and 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, $^{238}\text{U} = 0.4 \mu\text{g/l}$, $^{236}\text{U}/^{238}\text{U}$ atomic ratio = 6×10^{-8} and $^{236}\text{U} = 6 \times 10^7 \text{ atom/L}$.

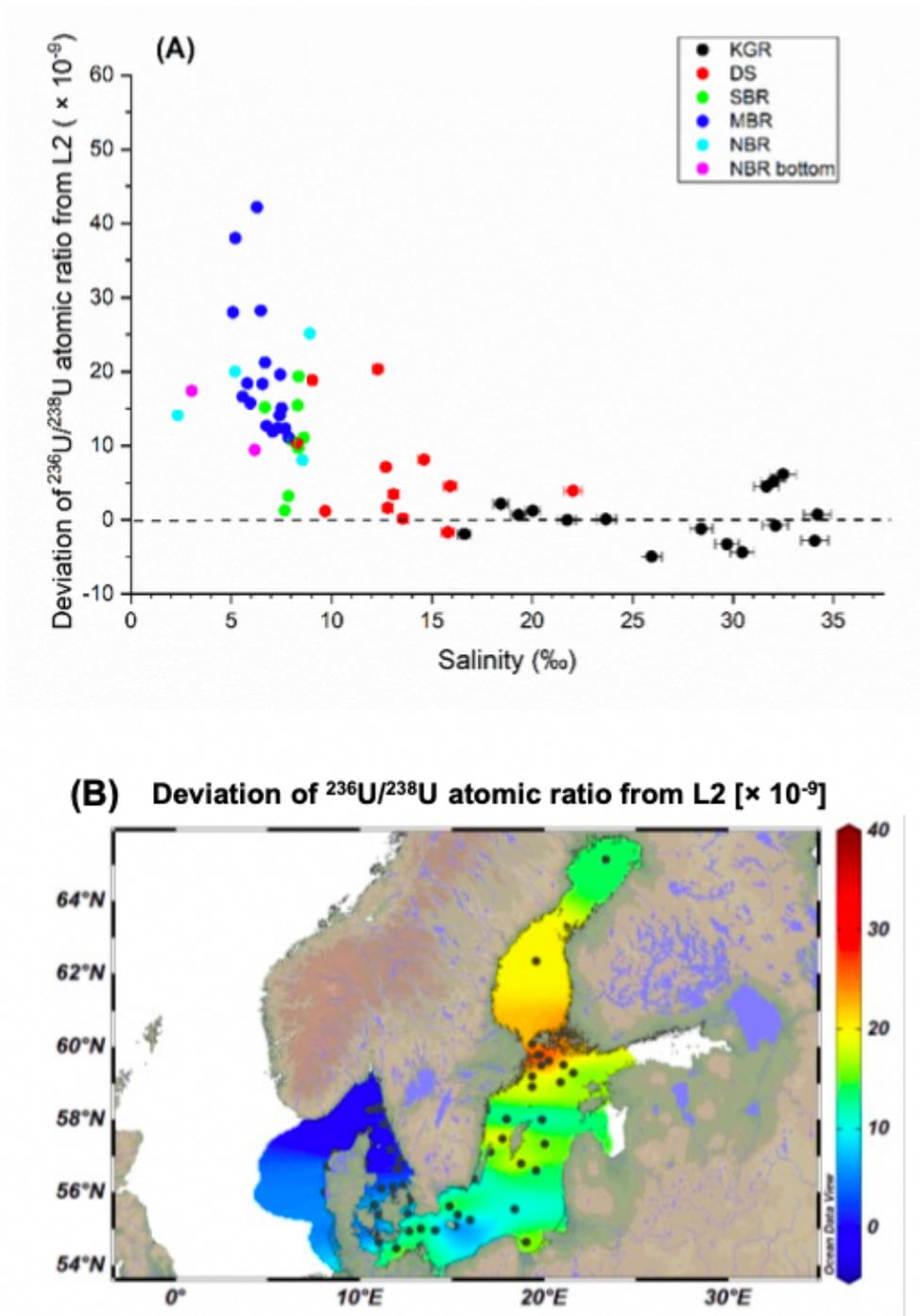


Figure 6

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) (L2 refers to the red dashed line in Figure 4A)

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