



Anthropogenic ^{236}U and ^{233}U in the Baltic Sea: Distributions, source terms, and budgets

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ABSTRACT

The Baltic Sea receives substantial amounts of hazardous substances and nutrients, which accumulate for decades and persistently impair the Baltic ecosystems. With long half-lives and high solubility, anthropogenic uranium isotopes (^{236}U and ^{233}U) are ideal tracers to depict the ocean dynamics in the Baltic Sea and the associated impacts on the fates of contaminants. However, their applications in the Baltic Sea are hampered by the inadequate source-term information. This study reports the first three-dimensional distributions of ^{236}U and ^{233}U in the Baltic Sea (2018–2019) and the first long-term hindcast simulation for reprocessing-derived ^{236}U dispersion in the North-Baltic Sea (1971–2018). Using $^{233}\text{U}/^{236}\text{U}$ fingerprints, we distinguish ^{236}U from the nuclear weapon testing and civil nuclear industries, which have comparable contributions (142 ± 13 and 174 ± 40 g) to the ^{236}U inventory in modern Baltic seawater. Budget calculations for ^{236}U inputs since the 1950s indicate that, the major ^{236}U sources in the Baltic Sea are the atmospheric fallouts (~ 1.35 kg) and discharges from nuclear reprocessing plants (> 211 g), and there is a continuous sink of ^{236}U to the anoxic sediments (589 ± 43 g). Our findings also indicate that the limited water renewal endows the Baltic Sea a strong "memory effect" retaining aged ^{236}U signals, and the previously unknown ^{236}U in the Baltic Sea is likely attributed to the retention of the mid-1990s' discharges from the nuclear reprocessing plants. Our preliminary results demonstrate the power of ^{236}U - ^{129}I dual-tracer in investigating water-mass mixing and estimating water age in the Baltic Sea, and this work provides fundamental knowledge for future ^{236}U tracer studies in the Baltic Sea.

1. Introduction

The Baltic Sea is a semi-enclosed marginal sea in Northern Europe comprised of a series of shallow sub-basins divided by sills. The inputs of freshwater from the vast catchment and saline water from the North Sea result in a density-stratified water structure (Burchard et al., 2005; Meier, 2007; Mohrholz et al., 2015). Limited water exchange and ~ 30 -year water residence time (Franck et al., 1987) make the Baltic Sea vulnerable to anthropogenic disturbances. Via river runoff and

atmospheric deposition, substantial amounts of hazardous substances and nutrients enter the Baltic Sea and accumulate for decades (HELCOM, 2018a) through the discharges from the highly populated (~ 85 million residents) catchment areas. Interactions between natural variability, anthropogenic pressures, and climate change lead to stronger stratification, less O_2 -rich saline inflows, and increasing eutrophication and deoxygenation of bottom waters since the 1980s (Carstensen et al., 2014; HELCOM, 2021).

Slow water circulation plays a significant role in determining the

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fates of nutrients/pollutants in the Baltic Sea (HELCOM, 2018b). Consequently, efforts have been dedicated to using the radionuclides (^{137}Cs , ^{99}Tc , ^{129}I , and ^{236}U), namely, those discharged from the two European nuclear reprocessing plants at Sellafield and La Hague, as oceanic tracers to track the formation, transport, and mixing of water masses in the Baltic Sea (Yi et al., 2011; Qiao et al., 2017, 2020a, 2021). Compared with the commonly used tracers in the Baltic Sea including salinity, temperature, nutrients, and dissolved organic matters (Aarup et al., 1996; Højerslev et al., 1996; Aure, 1998; Meier, 2005, 2007; Stedmon et al., 2010; Kristiansen and Aas, 2015), the reprocessing-derived radionuclides are characterized by extremely low natural backgrounds, nonequilibrium dispersion states in the ocean, and accessible discharge histories. In particular, their well-constrained point-source releases are favorable for the hindcast simulations with reliable input functions to depict the large-scale and long-term hydrodynamics in the Baltic Sea.

Among the reprocessing-derived radionuclides, ^{129}I ($t_{1/2}=15.7$ Ma) and ^{236}U ($t_{1/2}=23.4$ Ma) are promising candidates with long radioactive half-lives (i.e. no decay correction needed) and high solubility. In addition, the reverse temporal discharging trends from the reprocessing plants between ^{129}I and ^{236}U lead to a monotonic increase of reprocessing-derived $^{129}\text{I}/^{236}\text{U}$ atomic ratios in the North Sea since the 1970s (Christl et al., 2015c), making $^{129}\text{I}/^{236}\text{U}$ atomic ratio an ideal chronologic tracer. The combination of ^{129}I and ^{236}U has been used to quantify the circulation timescale of Atlantic waters in the Arctic Ocean (Casacuberta et al., 2016; Wefing et al., 2021) and can be potentially applied for estimating the ages of the saline inflowing waters from the North Sea to the Baltic Sea, which were only calculated by three-dimensional (3D) ocean models (Meier, 2007, 2005) but not verified by any observation data. Up to now, there have been numerous works about the sources, distributions, speciation, transport, and tracer applications of ^{129}I in the Baltic Sea (Hou et al., 2002, 2007; Hansen et al., 2011; Yi et al., 2013a, 2013b, 2011), but little information is known about ^{236}U .

During the Atomic Era, significant amounts of ^{236}U have been released into the environment by human nuclear activities. ^{236}U is mainly produced by thermal neutron capture reaction $^{235}\text{U}(\text{n}, \gamma)^{236}\text{U}$ in nuclear reactors (NRs) and fast neutron reaction $^{238}\text{U}(\text{n}, 3\text{n})^{236}\text{U}$ in nuclear explosions (Steier et al., 2008; Sakaguchi et al., 2009). As a result, the two primary sources of anthropogenic ^{236}U in the environment are global fallout (GF) from atmospheric nuclear weapons testing (900 - 2100 kg; Sakaguchi et al., 2009; Christl et al., 2012; Winkler et al., 2012; Casacuberta et al., 2014) and the authorized discharges from the reprocessing plants (> 260 kg; Castrillejo et al., 2020). However, the ubiquitous GF-derived ^{236}U interferes with the point-source-released reprocessing-derived ^{236}U in observations (Christl et al., 2015a). Until recently the accessible measurement of another anthropogenic uranium isotope, ^{233}U ($t_{1/2}=0.16$ Ma), in environmental samples by accelerator mass spectrometry provided a new possibility to discriminate ^{236}U sources between GF and NR (Hain et al., 2020). ^{233}U is primarily produced by thermal neutron capture reaction $^{232}\text{Th}(\text{n}, \gamma)^{233}\text{U}$ in some thorium-based prototype reactors and fast neutron reaction $^{235}\text{U}(\text{n}, 3\text{n})^{233}\text{U}$ in nuclear explosions (Hain et al., 2020). In contrast to ^{236}U , almost no ^{233}U is produced in commercial NRs and released by reprocessing plants, and the only substantial release of ^{233}U (7 - 15 kg; Lin et al., 2021b) is from the tests of ^{233}U -fueled devices and thermonuclear weapons using enriched uranium as tamper material (Hain et al., 2020). This difference makes the $^{233}\text{U}/^{236}\text{U}$ atomic ratio a source-specific fingerprint allowing possible distinction between different emission sources for ^{236}U (Hain et al., 2020; Qiao et al., 2020b, 2021; Lin et al., 2021b, 2021c). The representative $^{233}\text{U}/^{236}\text{U}$ atomic ratio for the integrated GF signal was suggested to be $(1.40 \pm 0.15) \times 10^{-2}$ by recent research on a peat core (Black Forest, Germany) and a coral core (Kume Island, Japan) (Hain et al., 2020). The reactor-modeling result and documented La Hague discharge data indicate that the $^{233}\text{U}/^{236}\text{U}$ atomic ratio in the NR-related releases from normal operations and accidents

should be at the level of 10^{-8} - 10^{-6} (Naegeli, 2004; Hain et al., 2020; HELCOM, 2020). This is supported by relatively low $^{233}\text{U}/^{236}\text{U}$ atomic ratios (avg. $(0.12 \pm 0.01) \times 10^{-2}$) measured in the seawater and sediments close to Sellafield (Hain et al., 2020). Using a binary end-member mixing algorithm, the GF-derived and NR-derived ^{236}U signals can be quantified in a multiple-source environment (e.g. the Baltic Sea and the Greenland coast; Hain et al., 2020; Qiao et al., 2020b, 2021; Lin et al., 2021b), and thus facilitating their tracer applications in the ocean.

Due to the limitations of measurement techniques in the past, observations of ^{236}U and ^{233}U in the Baltic Sea are scarce. Qiao et al. (2017) and Lin et al. (2021b) investigated the levels and distribution of ^{236}U in the Danish coastal waters and temporal evolution of ^{236}U and ^{233}U in the central Baltic Sea, respectively. They demonstrated that the Baltic Sea receives ^{236}U from both GF and reprocessing plants. A follow-up study by Qiao et al. (2021) reported the distributions of ^{236}U and ^{233}U in the surface waters of the Baltic Sea and suggested an "unknown" local NR-derived ^{236}U source, e.g. the undocumented discharges from local nuclear facilities or accidental leakages from nuclear waste dumping. However, there are still many knowledge gaps of the ^{236}U and ^{233}U in the Baltic Sea, e.g. their 3D distributions, major (input/output) fluxes, and budgets in seawater. In addition, pinpointing the source of the "unknown" NR-derived ^{236}U in the Baltic Sea requires further investigation to ease the public concern on radiological safety. Hence, systematic observations of ^{236}U and ^{233}U become the prerequisite of their tracer applications in the Baltic Sea.

In this work, we present the first dataset of 3D distribution of ^{236}U and ^{233}U in the modern Baltic Sea (2018–2019). Using the novel $^{233}\text{U}/^{236}\text{U}$ signature, we aim to locate the source terms of ^{236}U and quantify their contributions to the total ^{236}U budget in the Baltic seawater. By combining observation and numerical modeling, we also seek to uncover the transport and mixing processes related to the dispersion of NR-derived ^{236}U from the North Sea to the Baltic Sea, thus providing a better understanding of the dynamics and fates of contaminants in the Baltic Sea.

2. Material and methods

2.1. Seawater sampling

To investigate the spatial distributions of ^{236}U and ^{233}U in the Baltic Sea, 125 seawater samples were collected from 35 stations (depth files) in the Baltic Sea during 2018–2019 (Table 1). The scientific expeditions were carried out by the Swedish Meteorological and Hydrological Institute (SMHI), the National Institute of Aquatic Resources at Technical University of Denmark (DTU Aqua), the Finnish Environment Institute and Radiation and Nuclear Safety Authority (SYKE and STUK), and the Leibniz Institute for Baltic Sea Research Warnemünde (IOW), Germany. Based on the sampling periods and geographical distribution, these samples could be grouped into three transects: Transect #1 from the Skagerrak to Western Gotland Basin in the winter of 2018; Transect #2 from the Bothnian Bay to the Gulf of Finland in the summer of 2019; and Transect #3 from the Arkona Basin to the Western Gotland Basin in the winter of 2019 (Fig. 1). The sampling area covers all major regions of the Baltic Sea, including the transition zone between the North Sea and Baltic Sea (Skagerrak, Kattegat, and Danish Straits), the southern Baltic Sea region (Arkona Basin, Bornholm Basin, and South Baltic Proper), the middle Baltic Sea region (Northern Baltic Proper, Western Gotland Basin, and Eastern Gotland Basin), and the northern Baltic Sea region (Archipelago Sea, Åland Sea, Bothnian Sea, Bothnian Bay, and Gulf of Finland).

To facilitate the presentation of results and discussions, we identified six water masses in the Baltic Sea based on the salinity and geographical locations. In the transition zone, the Atlantic water (salinity > 34.5), Jutland coastal water (salinity = 32 - 34.5), and Baltic inflowing/outflowing water (salinity = 15 - 32) are three dominating water masses (Aarup et al., 1996). The Atlantic water dominates the bottom of

Table 1

Description of scientific expeditions and sampling information.

Transect no.	Cruise	Principal	Survey period	Vessel	Station number	Sample number
1	October monitoring cruise	SMHI	2018-10-14 - 2018-10-21	R/V Aranda	5	8
1	November monitoring cruise	SMHI	2018-11-08 - 2018-11-15	R/V Aranda	7	10
1	December monitoring cruise	SMHI	2018-12-04 - 2018-12-11	R/V Aranda	1	3
1	Cruise 11/2018	DTU Aqua	2018-11-03 - 2018-11-14	R/V Dana	6	24
2	COMBINE 2	SYKE SYUK	2019-05-27 - 2019-06-07	R/V Aranda	7	16
3	EMB-224	IOW	2019-10-11 - 2019-10-23	R/V Elisabeth Mann Borgese	13	64

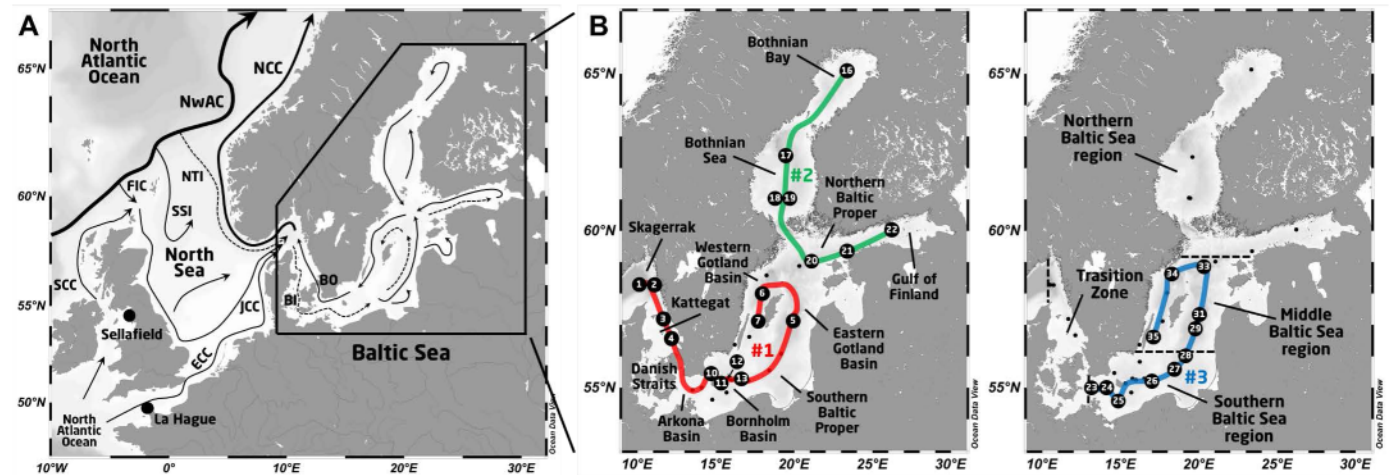


Fig. 1. (A) Schematic circulation patterns in the North Sea-Baltic Sea region; (B) three transects and sampling stations in the Baltic Sea. In plot A, the solid and dashed arrows represent surface and bottom currents, respectively. In plot B, the numbered black circles represent sampling stations; and the red, green, blue lines represent Transect #1 from the Skagerrak to the Western Gotland Basin, Transect #2 from the Bothnian Bay to the Gulf of Finland, and Transect #3 from the Arkona Basin to the Western Gotland Basin, respectively. The abbreviations in plot A are: NwAC, Norwegian Atlantic Current; NTI, Norwegian Trench Inflow; FIC, Fair Isle Current; SCC, Scottish Coastal Current; ECC, English Channel Current; JCC, Jutland Coastal Current; NCC, Norwegian Coastal Current; BI, Baltic Inflow; BO, Baltic Outflow.

Skagerrak and Kattegat and originates from a southeastward branch of the Norwegian Atlantic Current expanding along the slope of the Norwegian Trench (Andersson, 2007). The Jutland coastal water is comprised of the central/southern North Sea waters and the runoff-influenced English Channel water, which transports waters from the southern North Sea to the transition zone by the Jutland Coastal Current (Aure, 1998). The main body of the Baltic Sea is separated by a permanent halocline into the Baltic bottom water (salinity = 9–15) and Baltic surface water (salinity = 5–9), which are influenced by the saline inflows from the transition zone, less saline brackish water from the northern Baltic region, and large freshwater supply from the rivers. The Baltic freshwater (salinity < 5) is mainly distributed in the Bothnian Bay and the estuary of Neva River in the Gulf of Finland.

2.2. Determination of ^{238}U , ^{236}U , and ^{233}U

^{238}U concentrations in seawater samples were directly measured by inductively coupled plasma mass spectrometry (Agilent 8800 ICP-QQQ) after 20–50 times dilution. For the determination of ultra-trace levels of ^{236}U and ^{233}U , an optimized radiochemical separation procedure was utilized to extract uranium isotopes from 4.5–9 L seawater samples (Qiao et al., 2015; Lin et al., 2021c), which is detailed in Supporting Information. Procedure blanks (in a total of nineteen) were prepared in every batch (typically seven samples) of analysis using ultrapure water following the same analytical procedure. Special measures were implemented to reduce the backgrounds of ^{236}U and ^{233}U , including purification of chemicals, utilization of laminar flow bench, and acid boiling for glassware (Lin et al., 2021c). Less than 1.95 ng of ^{238}U , 2.79×10^6 atoms of ^{236}U , and 8.94×10^3 atoms of ^{233}U were detected in the procedure blanks, which were negligible compared to the uranium

isotopes measured in our seawater samples. The accelerator mass spectrometry measurements of the atomic ratios of $^{236}\text{U}/^{238}\text{U}$ and $^{233}\text{U}/^{238}\text{U}$ were carried out at the Vienna Environmental Research Accelerator (VERA) facility in the University of Vienna using the established setup and method for actinides (Steier et al., 2019; Hain et al., 2020;). ^{238}U was also used as an intrinsic tracer to obtain the chemical yield of uranium, which was on an average of ~70% in this work. The robustness of the analytical method was verified by repeatedly analyzing a quality control sample (2018184, Baltic seawater), provided by the Federal Maritime and Hydrographic Agency of Germany, together with the seawater samples in this work (Lin et al., 2021c). The background subtraction and uncertainty calculation for the actual uranium concentrations and isotope ratios are detailed in Supporting Information.

2.3. Hindcast simulation

In this work, a 3D ocean circulation model HIROMB-BOOS Model (HBM) was applied to simulate the dispersion of reprocessing-derived ^{236}U in the North-Baltic Sea. More details of the HBM model, setup, simulation, and validation are given in Supporting Information.

HBM is a well-calibrated operational model for storm surge forecast, oil spill forecast warning service in Denmark and Germany (Berg and Poulsen, 2012; She and Murawski, 2018; Murawski et al., 2021). The model has three nested layers: (1) a 2D barotropic model covering the North Atlantic Shelf Sea in 6-nautical-mile resolution to provide wind-induced sea level for the 3D regional model; (2) a 3D North-Baltic Sea model in 3-nautical-mile resolution, which is two-way nested with two high-resolution sub-domains; (3) two 0.5-nautical-mile model sub-domains in the Wadden Sea and the transition zone between the

North Sea and the Baltic Sea. The dynamic two-way nesting with high resolution is essential for resolving the narrow Danish Straits and simulating the water exchange between the North Sea and the Baltic Sea (She et al., 2007).

Two point sources were set for the two European reprocessing plants at La Hague and Sellafield. The annual discharges of ^{236}U from two point sources were based on the reconstructed data from shell records by Castrillejo et al. (2020), and a constant discharge rate was assumed for each point source within a specific year. As Sellafield is located out of the North-Baltic Sea domain, the corresponding point source was moved to the upper left corner of the domain. A loss rate of 50% and a lag time of 1 year were assumed for the transport of ^{236}U from Sellafield to its new location as suggested by Christl et al. (2015b). The model was simulated from 1971 to 2018 and validated with the observed NR-derived ^{236}U concentrations in Transect #1.

3. Results

3.1. Levels and distributions of ^{238}U , ^{236}U , ^{233}U concentrations and $^{236}\text{U}/^{238}\text{U}$, $^{233}\text{U}/^{238}\text{U}$ atomic ratios

An overview of the concentrations of ^{238}U , ^{236}U , and ^{233}U , as well as the atomic ratios of $^{236}\text{U}/^{238}\text{U}$, $^{233}\text{U}/^{238}\text{U}$, and $^{233}\text{U}/^{236}\text{U}$ in the Baltic Sea in 2018–2019 is given in Table S1. The ^{238}U concentrations ranged between 0.3 and 3.8 $\mu\text{g}/\text{kg}$ and showed a strong correlation with salinity ($R^2=0.999$, Fig. 2A) in the surface waters (depth < 10 m) of the Baltic Sea. The intercept of linear fitting suggested an average riverine ^{238}U concentration of 0.06 $\mu\text{g}/\text{kg}$. However, compared to the calculated ^{238}U concentrations from the surface ^{238}U -salinity correlation, negative offsets were apparent (avg. -15%) for measured ^{238}U concentrations in the sulfidic deep waters of the central Baltic Sea, and the offsets showed a strong connection with sulfide concentrations (Fig. 2B and C).

The measured $^{236}\text{U}/^{238}\text{U}$ and $^{233}\text{U}/^{238}\text{U}$ atomic ratios ($(2.42 - 25.8) \times 10^{-9}$ and $(1.42 - 22.5) \times 10^{-11}$, respectively) in the Baltic seawater collected in 2018–2019 were significantly higher than their natural background ratios of $10^{-14} - 10^{-10}$ and $10^{-14} - 10^{-11}$, respectively (Steier et al., 2008; Hain et al., 2020), suggesting the dominance of

anthropogenic ^{236}U and ^{233}U in the Baltic Sea. $^{236}\text{U}/^{238}\text{U}$ and $^{233}\text{U}/^{238}\text{U}$ atomic ratios in this work were comparable to the reported values in Baltic surface waters in 2014–2015 ($(5.47 - 52.4) \times 10^{-9}$ and $(0.77 - 28.6) \times 10^{-11}$, respectively; Qiao et al., 2021). The obtained $^{236}\text{U}/^{238}\text{U}$ ($(14.8 - 16.6) \times 10^{-9}$) and $^{233}\text{U}/^{238}\text{U}$ ($(7.60 - 9.82) \times 10^{-11}$) atomic ratios in the bottom waters of the Gotland Basin and the Landsort Deep were also consistent with the values ($(14.5 - 16.3) \times 10^{-9}$ and $(7.54 - 10.8) \times 10^{-11}$, respectively) previously determined in the ~2018 segments of sediment cores collected at the same sites (Lin et al., 2021b).

The measured ^{236}U ($(1.99 - 8.75) \times 10^7$ atoms/kg) and ^{233}U ($(1.31 - 3.73) \times 10^5$ atoms/kg) concentrations in this study were also similar to the previously determined levels in the surface waters of the Baltic Sea in 2011–2016 ($(3.31 - 12.5) \times 10^7$ atoms/kg and $(0.46 - 5.63) \times 10^5$ atoms/kg, respectively; Qiao et al., 2017, 2021). A comparison of currently available ^{236}U datasets (Fig. 3) indicated that the North Sea and the Baltic Sea have higher ^{236}U concentrations than the inflowing eastern North Atlantic Central waters to the North Sea. As the eastern North Atlantic Central waters solely carry GF signal (Castrillejo et al., 2018), the elevated ^{236}U concentrations in the North Sea and the Baltic Sea should be attributed to the regional sources (i.e. reprocessing plants). The ^{236}U concentrations in the Baltic Sea were slightly lower than those in the North Sea, and relatively high ^{236}U concentrations ($> 5 \times 10^7$ atoms/kg) were observed in the Jutland coastal water, Baltic inflowing/outflowing water, and part of Baltic bottom water along the passage from the Danish Straits to the Gotland Basin (Fig. 4), suggesting the transport of ^{236}U from the North Sea to the Baltic Sea. The ^{233}U concentrations were distributed within a narrow range ($(2.15 - 3.50) \times 10^5$ atoms/kg) throughout the Baltic Sea with slightly lower levels in the Atlantic water and Baltic freshwater.

3.2. Levels and distributions of $^{233}\text{U}/^{236}\text{U}$ atomic ratios, GF-derived and NR-derived ^{236}U concentrations

The atomic ratios of $^{233}\text{U}/^{236}\text{U}$ in the Baltic Sea were within $(0.18 - 0.97) \times 10^{-2}$ in 2018–2019, reflecting contributions of ^{236}U from both GF and NR sources. Higher $^{233}\text{U}/^{236}\text{U}$ atomic ratios as observed in the Bothnian Bay indicated higher relative contributions of GF-derived ^{236}U ,

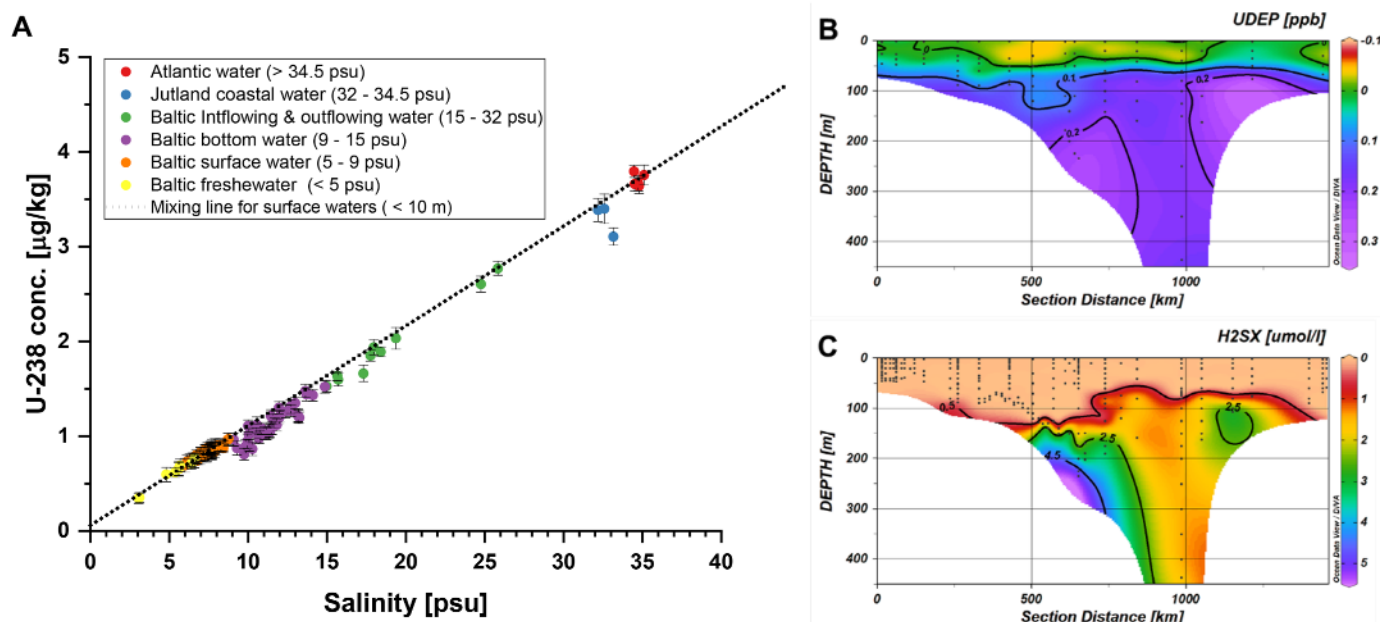


Fig. 2. The mixing diagram of ^{238}U concentration vs salinity in the Baltic Sea in 2018–2019 (A) and the distribution of uranium depletion (B) and sulfide concentrations (C) in Transect #3. In plot A, the mixing line function for the surface waters (depth < 10 m) is $C^{238} = 0.105 \times S + 0.0587$ ($R^2 = 0.999$), where C^{238} and S represent the ^{238}U concentration ($\mu\text{g}/\text{kg}$) and salinity (PSU), respectively. In plot B, uranium depletion is defined as the difference between the salinity-based ^{238}U concentration calculated by the aforementioned function and the measured ^{238}U concentration in the seawater.

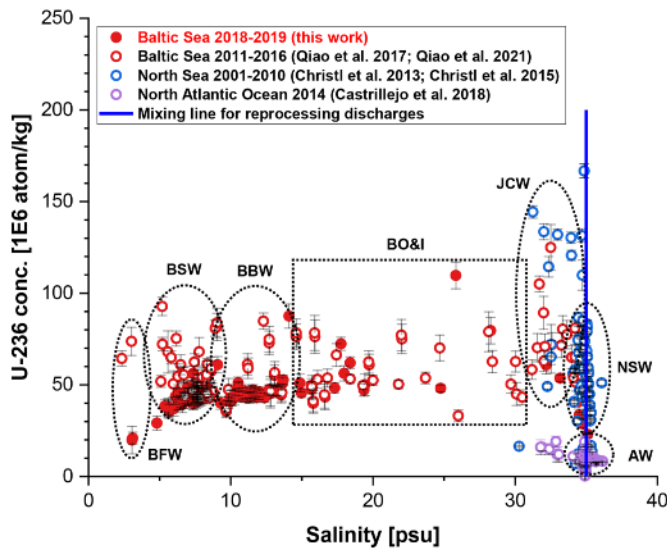


Fig. 3. Comparison with currently available ^{236}U datasets in the Baltic Sea, North Sea, and North Atlantic Ocean. Red open and solid circles represent the Baltic Sea waters sampled in 2011–2016 (Qiao et al., 2017; Qiao et al., 2021) and 2018–2019 (this work); blue open circles represent the North Sea waters sampled in 2001–2010 (Christl et al., 2015a, 2017); and purple open circles represent the eastern North Atlantic Central water sampled in 2014 (Castrillejo et al., 2018). The blue line represents a binary mixing between the anthropogenic ^{236}U signals (global fallout and reprocessing plant discharges) and negligible natural ^{236}U signals in the open ocean. The abbreviations in the plot are: AW, Atlantic water; NSW, North Sea water; JCW, Jutland coastal water; BO&I, Baltic outflowing and inflowing waters; BBW, Baltic bottom water; BSW, Baltic surface water; BFW, Baltic freshwater.

whereas lower $^{233}\text{U}/^{236}\text{U}$ atomic ratios in the Jutland coastal water, Baltic inflow water, and Baltic bottom water reflected the relatively strong influences of NR-derived ^{236}U in the transition zone and southern Baltic region (Fig. 5).

Using $^{233}\text{U}/^{236}\text{U}$ atomic ratio in a binary mixing algorithm consisting of GF-derived ^{236}U ($^{233}\text{U}/^{236}\text{U} = 1.40 \times 10^{-2}$) and NR-derived ^{236}U ($^{233}\text{U}/^{236}\text{U} = 10^{-6}$) as endmembers, the GF-derived and NR-derived ^{236}U concentrations in all seawater samples were calculated (see details in Supporting Information), which varied within $(0.93 - 2.66) \times 10^7$ and $(0.61 - 6.90) \times 10^7$ atoms/kg, respectively (Fig. 5). In the brackish waters of the Baltic Sea (Baltic inflowing/outflowing water, Baltic surface water, and Baltic bottom water), the GF-derived ^{236}U concentrations $((1.48 - 2.66) \times 10^7$ atoms/kg) were evenly distributed, whereas the freshwater and saline sources (Atlantic water and Jutland coastal water) had slightly lower GF-derived ^{236}U concentrations $((0.93 - 1.66) \times 10^7$ atoms/kg). The GF-derived ^{236}U concentrations $((1.8 - 2.5) \times 10^7$ atoms/kg) in the surface waters of the central Baltic Sea in 2018–2019 were consistent with the reported values $((1.2 - 3.9) \times 10^7$ atoms/kg) in 2011–2016 (Qiao et al., 2021). However, these concentration values were much higher than the representative GF levels in the surface water of the open ocean (0.8×10^7 atoms/kg; Christl et al., 2012) and simulated concentrations in the North Sea in the 2010s $((0.7 - 1.1) \times 10^7$ atoms/kg; Christl et al., 2015b) derived from a global surface ocean model. Higher NR-derived ^{236}U concentrations $((4.21 - 9.59) \times 10^7$ atoms/kg) mainly occurred in the sub-surface layer of the Skagerrak and Kattegat as well as the bottom inflows along transects from the Danish Straits to the northern Baltic proper. The lowest NR-derived ^{236}U concentrations $((0.61 - 1.31) \times 10^7$ atoms/kg) were observed in the Bothnian Bay and the bottom of Skagerrak predominated by the Baltic freshwater and Atlantic water, respectively.

3.3. Inventories of ^{236}U and ^{233}U in the modern baltic sea

Based on the observation data in 2018–2019, we extrapolated the

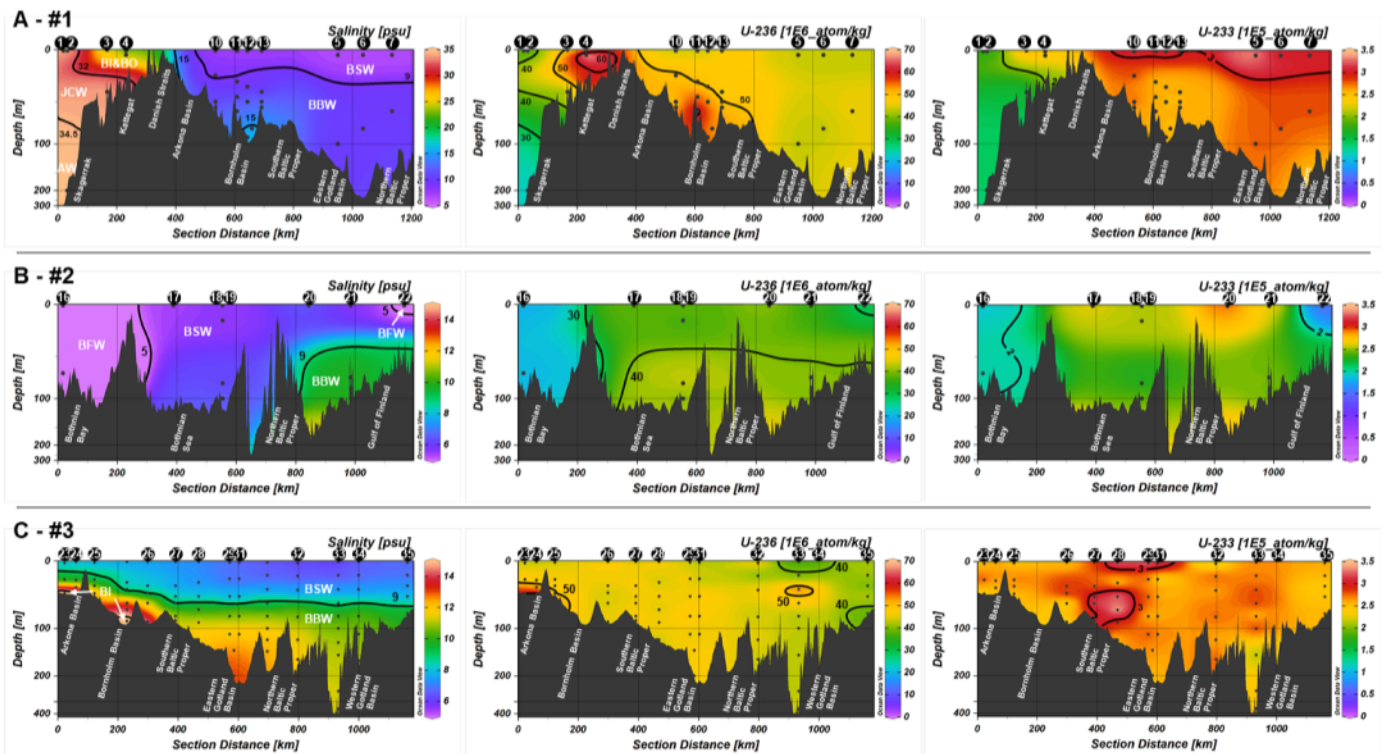


Fig. 4. Distributions of salinity, ^{236}U and ^{233}U concentrations in Transect #1 (A), Transect #2 (B), and Transect #3 (C). The numbered black circles in the plots represent sampling stations. The abbreviations in the plots are: AW, Atlantic water; JCW, Jutland coastal water; BI and BO, Baltic inflowing and outflowing waters; BBW, Baltic bottom water; BSW, Baltic surface water; BFW, Baltic freshwater.

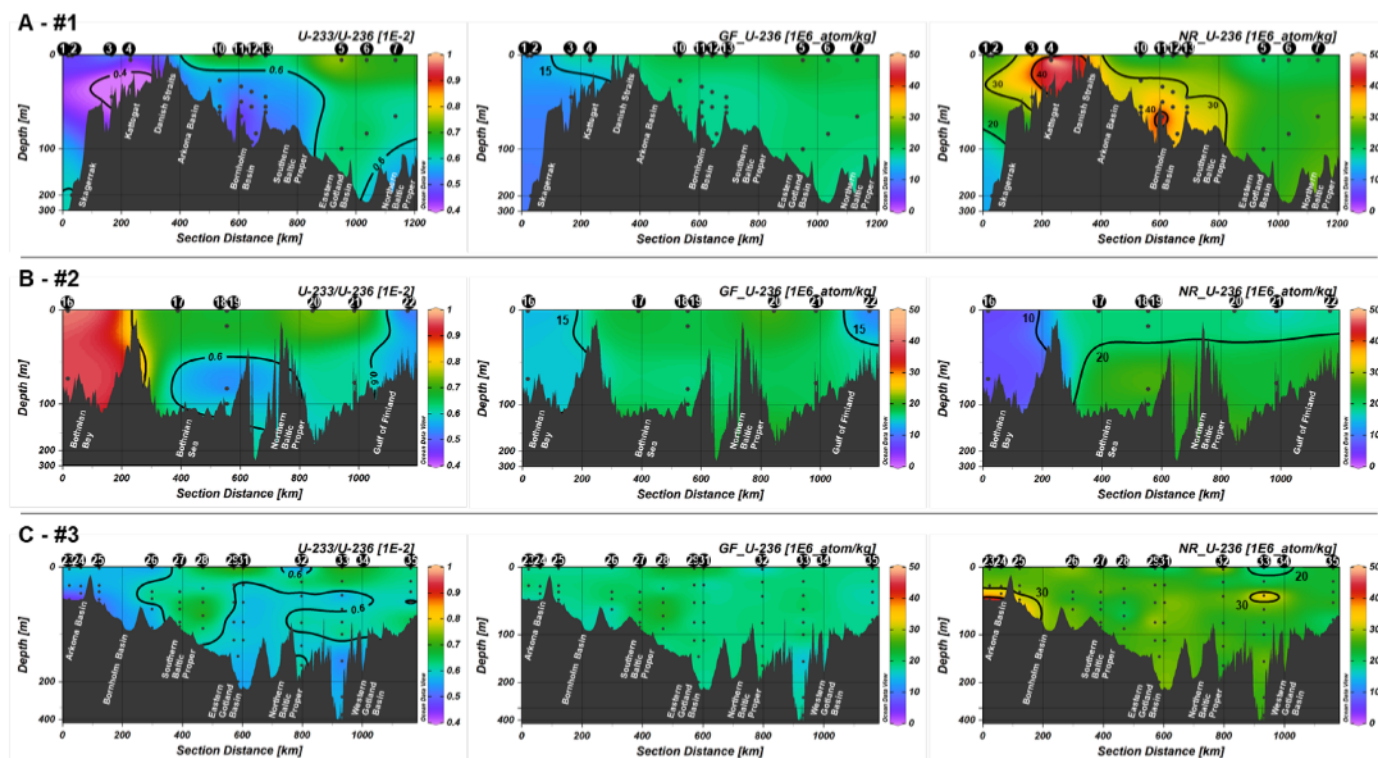


Fig. 5. Distributions of $^{236}\text{U}/^{238}\text{U}$ atomic ratios, GF-derived and NR-derived ^{236}U concentrations in Transect #1 (A), Transect #2 (B), and Transect #3 (C). The numbered black circles in the plots represent sampling stations. The abbreviations in the plots are: GF, global fallout; NR, nuclear reactor.

spatial distributions of ^{236}U and ^{233}U concentrations, as well as GF-derived and NR-derived ^{236}U concentrations, throughout the Baltic Sea using a 3D estimation tool in Ocean Data View (5.3.0). The 3D estimation was implemented as a fast weighted averaging procedure and used a specified high-resolution topography dataset of the Baltic Sea (latitude: 1° ; longitude: 2° ; depth: 5 m; Seifert et al., 2001). The total dissolved ^{236}U and ^{233}U inventories in the modern Baltic seawater were thereby estimated to be 316 ± 25 g and 1.96 ± 0.25 g, respectively. The GF-derived and NR-derived ^{236}U accounted for $44.9 \pm 4.2\%$ (142 ± 13 g) and $55.1 \pm 13.0\%$ (174 ± 40 g) of the total ^{236}U in the Baltic Sea, respectively.

4. Discussions

4.1. Behaviors of U in the Baltic Sea

The mixing diagram of ^{236}U concentration vs salinity (Fig. 2A) indicates that uranium has conservative behaviors in the oxic waters (e.g. surface waters) of the Baltic Sea, where uranium is present in +VI oxidation state as highly soluble uranyl carbonate complexes (Djogić et al., 1986). However, in the middle and northern Baltic Sea regions, 10 - 20% of ^{236}U was scavenged from the sulfidic deep waters in 2018–2019, and this fraction could be up to 40% reported by Anderson et al. (1989) and Dellwig et al. (2021). The Baltic bottom water is permanently hypoxic and periodically anoxic, as its renewal only relies on the irregularly strong barotropic inflow events, or so-called Major Baltic Inflows (Carstensen et al., 2014; Meier et al., 2019). The recent investigations on the water column and sediments of the central Baltic Sea (Dellwig et al. 2021; Lin et al., 2021b) suggested that the uranium depletion in sulfidic waters arises from the redox-driven processes in the water column and diagenesis-driven removal in the sediment: (1) the soluble U(VI) in the water column can be reduced to insoluble U(IV) by sulfides and deposited on the seabed as particulate forms (Anderson et al., 1989); and (2) the dissolved uranium can be removed from the sulfidic porewaters into sediments, leading to a diffusion of dissolved

uranium from bottom waters to the porewaters (Barnes and Cochran, 1990; Klinkhammer and Palmer, 1991; Lovley et al., 1991). The Major Baltic Inflows can temporally oxygenate the euxinic Baltic bottom water, suppress the removal of uranium from the water column, and remobilize a part of the scavenged uranium in the surface sediments (20 - 50%), leading to a release of uranium from the uppermost porewaters to the water column (Dellwig et al. 2021). However, the remobilization of sedimentary uranium is quite limited and temporal, which slightly elevates the ^{238}U concentrations in the near-seabed waters (Dellwig et al. 2021). The uranium depletion recurs after the reestablishment of the bottom euxinia in the central Baltic Sea region (Dellwig et al. 2021).

The Baltic sediments in the anoxic sub-basins generally act as a net sink for natural and anthropogenic uranium isotopes. Our previous investigation on the Baltic sediments indicated that 589 ± 43 g of ^{236}U (GF-derived: 266 ± 5 g; NR-derived: 323 ± 43 g) were scavenged from the Baltic seawater to the sediments (Lin et al., 2021b). The comparable atomic ratios of $^{236}\text{U}/^{238}\text{U}$, $^{233}\text{U}/^{238}\text{U}$, and $^{233}\text{U}/^{236}\text{U}$ (Section 3.1) between the bottom waters and surface sediments in the central Baltic Sea revealed no significant isotopic fractionation for uranium during the scavenging processes. The $^{233}\text{U}/^{236}\text{U}$ atomic ratio is not affected by the scavenging processes and thus can still be used as a robust signature for tracing the water-mass movement and identifying emission sources of anthropogenic uranium (Hain et al., 2020; Qiao et al., 2020b).

4.2. Source terms of ^{236}U in the Baltic Sea

In addition to a negligible natural background, the majority of ^{236}U in the Baltic Sea originates from GF-related and NR-related sources and enters the Baltic Sea through various pathways. The GF-derived ^{236}U in the modern Baltic seawater is potentially from: (1) the dissolution of the directly deposited GF-derived ^{236}U on the Baltic Sea; (2) the riverine inputs of GF-derived ^{236}U deposited on the catchment; and (3) the oceanic transport of dissolved GF-derived ^{236}U in the North Sea via Baltic inflows. The NR-derived ^{236}U in the modern Baltic seawater may be attributed to: (1) the marine transport of reprocessing-derived ^{236}U

from the North Sea; (2) the dissolved ^{236}U fallouts from the Chernobyl accident, both in Baltic seawater and river waters from the catchment; and (3) unreported discharges from local nuclear facilities or accidental leakages from dumped nuclear wastes. In the following subsections, we will further discuss the specific sources/inputs and sinks/outputs of GF-derived and NR-derived ^{236}U , investigate their related transport and mixing processes, and perform the budget calculations for them respectively.

4.3. NR-derived ^{236}U in the Baltic Sea

4.3.1. Major inputs of NR-derived ^{236}U

Our recent study on the Baltic sediments indicated that the ^{236}U fallouts from the Chernobyl accident were very limited in the Baltic region, and little Chernobyl-derived ^{236}U was dissolved in the Baltic seawater (Lin et al., 2021b). Even though our earlier findings suggested a potential unaccounted NR-derived ^{236}U source in the Baltic Sea (Qiao et al., 2021), the observations in 2018–2019 did not demonstrate distinctly high NR-derived ^{236}U concentrations in the Baltic bottom water or freshwater, indicating no significant releases of NR-derived ^{236}U from the local nuclear facilities or dumped nuclear wastes to the modern Baltic Sea. Both the spatial distribution of NR-derived ^{236}U (Fig. 5) and the mixing diagram of NR-derived ^{236}U vs salinity (Fig. 6A) suggested that the NR-derived ^{236}U in the modern Baltic seawater should be mainly from the North Sea (more specifically, two reprocessing plants) rather than other riverine or benthonic sources.

4.3.2. Transport and mixing of NR-derived ^{236}U

The NR-derived ^{236}U vs salinity diagram revealed that there were three endmembers for NR-derived ^{236}U in the Baltic Sea (Fig. 6A). The first endmember was the saline Atlantic water characterized by low NR-derived ^{236}U concentrations ($< 2 \times 10^7$ atoms/kg). The second endmember was the slightly less saline water (Jutland coastal water) with high and variable NR-derived ^{236}U concentrations ($(3.5 - 16) \times 10^7$ atoms/kg) constrained by the upper and lower boundaries for the potential mixing lines (red and blue dot lines in Fig. 6A). The third endmember was the brackish Baltic surface water with relatively constant NR-derived ^{236}U concentrations ($(1.5 - 3.5) \times 10^7$ atoms/kg), likely

arising from the long residence time of saline water in the Baltic Sea.

Due to the limitations in sampling frequency and spatial resolution, the mixing diagram lost many details in the transition zone and southern Baltic Sea region. To elaborate further on the transport/mixing processes affecting NR-derived ^{236}U distribution, the 3D ocean model HBM was used to simulate the dispersion of ^{236}U discharges from Sellafield and La Hague. It should be noted that there are methodological difficulties in distinguishing NR-derived ^{236}U from the reprocessing plants and those from other civil nuclear facilities in the observations, but only the former ones were considered in our simulation. For a more accurate description, we prefer to use the term "reprocessing-derived ^{236}U " to represent "NR-derived ^{236}U from the reprocessing plants" in the simulation results.

The simulated water-column results for 128 stations in the North-Baltic Sea drew a full picture for the dispersion of reprocessing-derived ^{236}U by oceanic circulation (Fig. S1). In the North Sea, higher ^{236}U concentrations were restricted in the near-shore areas rather than the central and northern regions by the coastal current transports. ^{236}U from Sellafield and La Hague was advected to the southern North Sea by the Scottish Coastal Current and English Channel Current, respectively, and further to the Skagerrak along the western European coastline by Jutland Coastal Current, which agreed with the previous observations (Christl et al., 2017, 2015a). Upon arrival at the Skagerrak, the Jutland coastal water tagged by reprocessing-derived ^{236}U was sandwiched into the surface brackish outflowing water and the bottom saline Atlantic water due to the density stratification, which was also confirmed by the earlier findings (Aarup et al., 1996; Dahlgaard et al., 1995; Stedmon et al., 2010).

The numerical modeling resolved an open question related to the mixing of NR-derived ^{236}U : why the Jutland coastal water had a variable endmember of NR-derived ^{236}U concentration (Fig. 6A)? The Jutland coastal water is a combination of the central/southern North Sea water and the English Channel water (Aure, 1998), thereby its endmember is determined by: (1) the NR-derived ^{236}U concentrations in the central/southern North Sea water and English Channel water; and (2) the fractions of central/southern North Sea water and the English Channel water. The NR-derived ^{236}U concentrations in both central/southern North Sea water and English Channel water were kept relatively stable

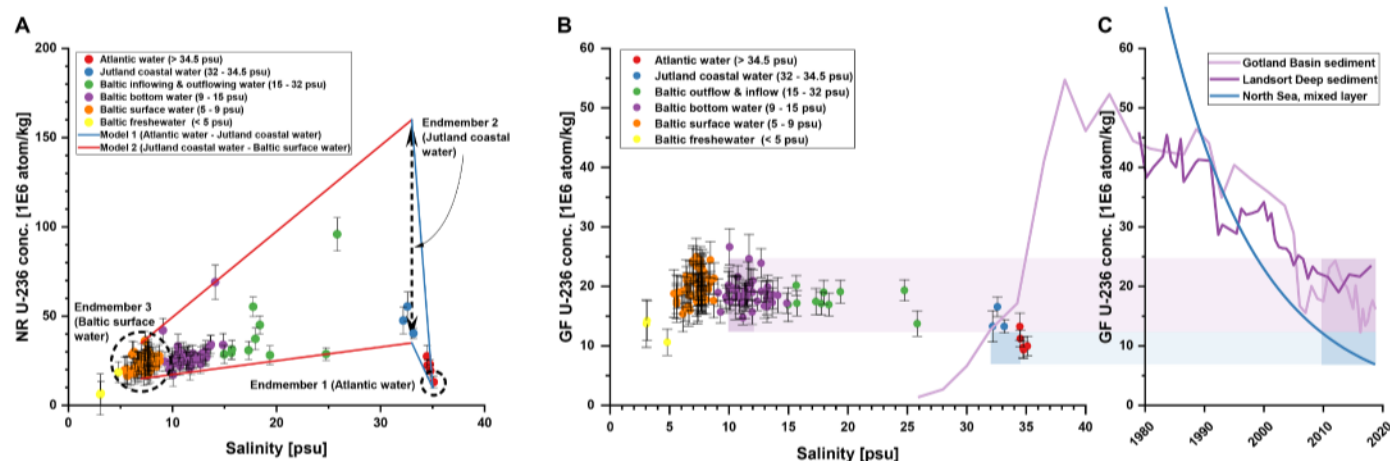


Fig. 6. The mixing diagrams of NR-derived ^{236}U concentration vs salinity (A) and GF-derived ^{236}U concentration vs salinity (B) in the Baltic Sea in 2018–2019, and a comparison of the resolved GF-derived ^{236}U concentrations in the bottom waters of the central Baltic Sea and the simulated GF-derived ^{236}U concentrations in the mixed layer of the North Sea (C). In plot A: the red solid lines represent the mixing lines between the Jutland coastal water and the Baltic surface water; the blue solid lines represent the mixing lines between the Atlantic water and the Jutland coastal water; the dashed circles and arrow line indicate the endmembers of three water masses. The three endmembers are summarized in Table S2, and the large variation in the second endmember is explained in Section 4.3.2). In plot B and plot C: the dark purple and blue rectangles represent the ranges of resolved and simulated GF-derived ^{236}U concentrations in the Baltic Sea and the North Sea in the 2010s, respectively. The resolved GF-derived ^{236}U concentrations are estimated from two sediment cores collected at the Gotland Basin and the Landsort Deep (Lin et al., 2021b). Here we assumed that the sedimentary GF-derived $^{236}\text{U}/^{238}\text{U}$ atomic ratios were equal to those in the bottom waters of two sub-basins and the ^{238}U concentrations are $1.2 \mu\text{g/kg}$ based on the historical bottom average salinity (ICES, 2020). The simulation data of GF-derived ^{236}U concentrations in the North Sea are from Christl et al. (2015b) and refer to the grid point 55°N , 5°E . The abbreviations in the plots are: GF, global fallout; NR, nuclear reactor.

during 2018–2019, as ^{236}U discharge rates from two reprocessing plants were generally constant since the 2010s. Our recent investigation on the water-mass compositions in the transition zone indicated that the fraction of the North Sea water has negligible inter-seasonal variability, whereas obvious seasonality is observed in the fraction of the English Channel water (Lin et al., 2021a). Driven by the predominant south-westerly winds in the North Sea, more English Channel water, along with La Hague-derived ^{236}U , flows into the Skagerrak in winter and vice versa in summer (Aure, 1998). The variations in the simulated reprocessing-derived ^{236}U concentrations ($(6 - 19) \times 10^7$ atoms/kg) of Jutland coastal water in 2018 were consistent with our observations ($(3.5 - 16) \times 10^7$ atoms/kg), and higher concentrations were observed in winter (Fig. 7A). In addition, both our observation and numerical modeling results revealed that the vertical mixings between the intermediate, less saline Jutland coastal water carrying relatively higher reprocessing signals and the bottom, more saline Atlantic water /surface, relatively fresh Baltic outflowing water in the transition zone caused the inverted v-shaped mixing lines.

4.3.3. Long-term dynamics of NR-derived ^{236}U

The long-term exchange of NR-derived ^{236}U between the North Sea and the Baltic was investigated by the temporal evolution of simulated inventories in both seas. As the ^{236}U discharges from the two reprocessing plants have decreased since the 1970s (Fig. 7C; Castrillejo et al., 2020), our hindcast simulation revealed a significant decline of reprocessing-derived ^{236}U concentrations in the Jutland coastal water

(Fig. 7B) and the decreasing inventories of NR-derived ^{236}U in the North Sea (Fig. 7D; from the max. 42.7 kg to 1.52 kg) and Baltic Sea (Fig. 7E; from the max. 624 g to 211 g) in recent 40 years. The decrease of NR-derived ^{236}U was much slower in the Baltic Sea than the North Sea, owing to a much longer water residence time in the Baltic Sea (~30 years) than the North Sea (1–2 years). The narrow and shallow channels of the Danish Straits restrict the bottom saline inflows driven by either the horizontal density gradients (observed in summer; Feistel et al., 2006) or storms (mainly in winter; Mohrholz, 2018). Limited water renewal endows the Baltic Sea with a strong "memory effect" and favors trapping the "aged" pollutions in the central Baltic Sea. The numerical modeling indicated that by the end of 2018, up to 57% of reprocessing-derived ^{236}U present in the Baltic Sea originated from the discharges before 1990. In contrast, almost 100% of reprocessing-derived ^{236}U in the North Sea was released within the recent 30 years. In the HBM model, even though the endmember for the Jutland coastal water varied largely for individual inflows, the monthly imported/exported reprocessing-derived ^{236}U was marginal (< 10%) compared to the NR-derived ^{236}U budget of the Baltic Sea since the 1990s, and could not significantly change the endmember for the Baltic surface water. This was also supported by our observations on the central Baltic Sea, where the distribution of NR-derived ^{236}U was relatively even (Figs. 5 and 6A).

Qiao et al. (2021) estimated that the "unknown" local source contributed 200 ± 47 g of NR-derived ^{236}U to the Baltic Sea, which was at the same level as the total NR-derived ^{236}U inventory obtained in this

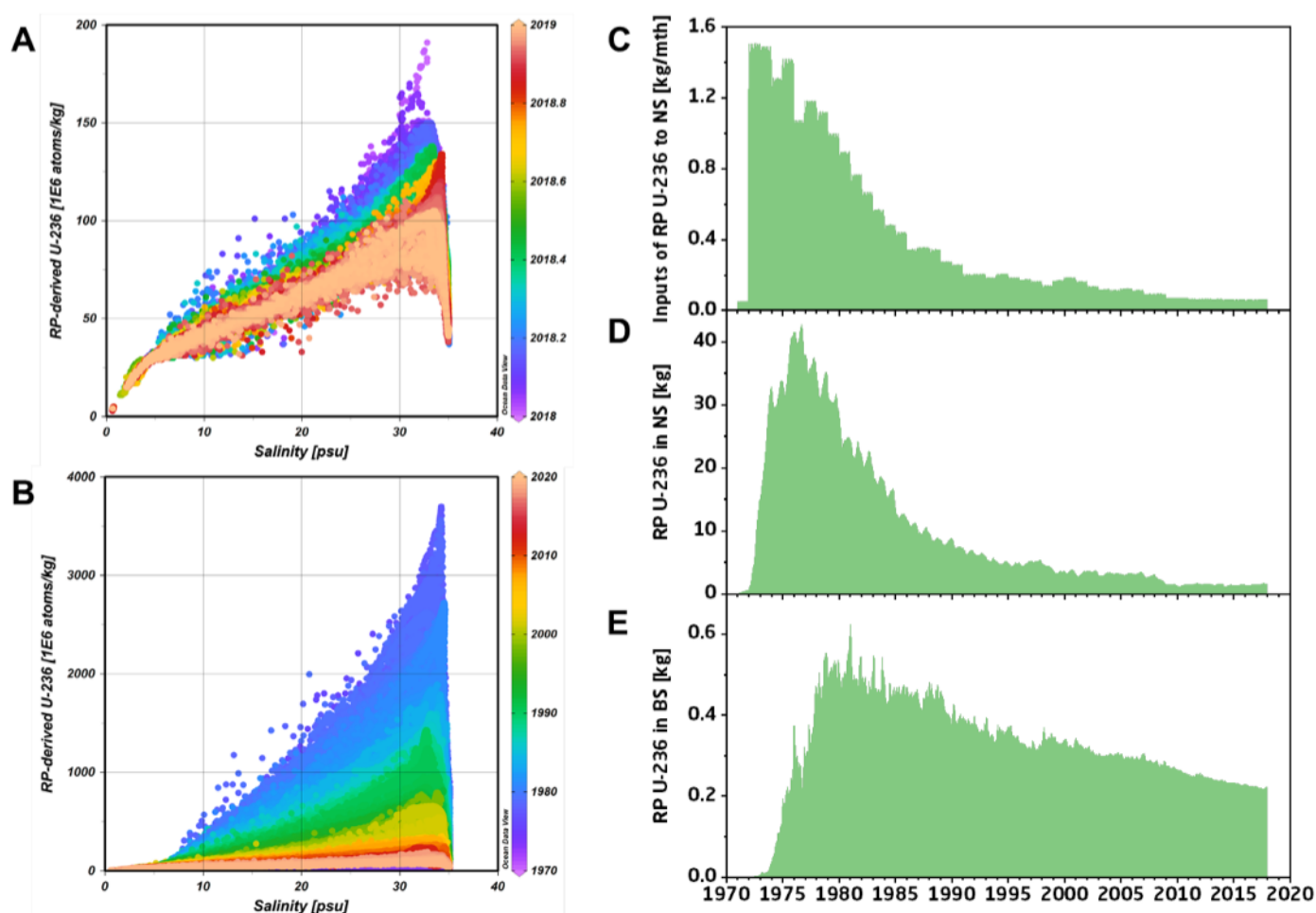


Fig. 7. Simulated mixing diagram of reprocessing-derived ^{236}U vs salinity for 74 stations in the transition zone and Baltic Sea in 2018 (A) and during 1971–2018 (B), monthly ^{236}U discharges from the two reprocessing plants to the North Sea in the hindcast simulation during 1971–2018 (C), and simulated inventories of reprocessing-derived ^{236}U in the North Sea (D) and Baltic Sea (E). The abbreviation "RP" in the plots refers to "reprocessing plant".

work. Considering the notable differences in sampling scales (periods, depths, and locations), endmember settings, and modeling approaches between the two studies (Fig. S2 and Table S2), the "unknown" source of NR-derived ^{236}U is likely the "aged" reprocessing-derived ^{236}U trapped in the central Baltic Sea. The direct evidence is the much lower ^{129}I /NR-derived ^{236}U atomic ratios in the surface waters of the middle Baltic Sea region (113 - 370) than those of the transition zone (1202 - 2608) in 2015 (Qiao et al., 2021). Our preliminary estimation suggested that the saline fraction of the surface waters of the central Baltic Sea carried at least 20 years older reprocessing signals than the saline waters of the transition zone (Fig. S3), which generally agreed with the simulated water-age results by Meier (2007). The endmember of Jutland coastal water for the "aged" reprocessing signal should be much higher than that for the "modern" signal, leading to the large deviation of ^{236}U / ^{238}U atomic ratios in the surface of the central Baltic Sea between the observations and the calculated results by the endmember mixing model adopted in our previous work (Qiao et al., 2021).

4.3.4. Budget calculation for NR-derived ^{236}U

Our budget calculation indicates a net input of 211 g of reprocessing-derived ^{236}U to the Baltic Sea, and 323 ± 43 g of NR-derived ^{236}U scavenged from the Baltic seawater to the sediments during 1971–2018 (Table 2). In general, there was a surplus of $(174 - (211 - 323) =) 286$ g for the observed inventory of NR-derived ^{236}U (174 ± 40 g) in the modern Baltic seawater, which most likely originate from the underestimated inputs from Sellafield due to the lack of its discharge history before 1970 (Castrillejo et al., 2020) and the undocumented discharges from the local nuclear facilities.

The overall ^{236}U budget scheme in the modern Baltic Sea (2018–2019) is shown in Fig. 8. The annual net output of NR-derived ^{236}U to the North Sea was estimated as 11 g/year (average value during 1981–2018 in hindcast simulation). Based on the investigation on the Baltic sediments, the scavenged NR-derived ^{236}U was estimated to be 5.3 g/year, assuming that the scavenging rate of NR-derived ^{236}U by the anoxic sediments was 2.7×10^{11} atom/($\text{m}^2 \cdot \text{year}$) in 2018 (Lin et al., 2021b) and an average hypoxic area of 49,000 km^2 in the Baltic Sea (Conley et al., 2009). Therefore, the Baltic Sea lost $5.3 + 11 = 16.3$ g of NR-derived ^{236}U in 2018. Considering that the anoxia in the middle Baltic Sea region will likely persist due to the large legacy of nutrients (HELCOM, 2018a), the loss of NR-derived ^{236}U in the Baltic Sea water will continue at least in this decade.

Table 2
The budget calculation for NR-derived ^{236}U in the Baltic Sea.

Period	Input/output for Baltic seawater [g] ¹			Seawater inventory [g]
	Oceanic transport ²	Local discharge ³	Sink to sedimen ⁴	
Before 1970	/	/	- 5 ± 2	/
1971 - 1980	565	/	- 34 ± 4	/
1981 - 1990	-177	/	- 90 ± 20	/
1991 - 2000	-53	/	- 59 ± 4	/
2001 - 2010	-83	0.40	- 97 ± 6	/
2011 - 2018	-41	0.04	- 39 ± 20	/
Overall	211	negligible	- 323 ± 43	174 ± 40
Current flux* [g/year]	-11	negligible	- 5.1 ± 2.9	- 16 ± 3

¹ Positive values represent the source/input, and negative values represent the sink/output; the slashes refer to "unknown".

² The fluxes of oceanic transport refer to the net fluxes taking all the inputs to/outputs from the Baltic Sea into account (see detailed explanations in Section 4.4.2).

³ The local discharges are from the HELCOM MORS discharge database (HELCOM, 2020).

⁴ The sink fluxes are estimated from two Baltic sediment cores (Lin et al., 2021b).

4.4. GF-derived ^{236}U in the Baltic Sea

4.4.1. Major inputs of GF-derived ^{236}U

The mixing diagram of GF-derived ^{236}U vs salinity (Fig. 6B) showed slightly higher GF-derived ^{236}U concentrations in the brackish waters of the Baltic Sea (Baltic surface water, Baltic bottom water, and Baltic outflow water) than the incoming saline and fresh waters (Atlantic water, Jutland coastal water, and Baltic freshwater). This phenomenon suggested that the present GF-derived ^{236}U in the main body of the Baltic Sea did not originate from the current inflowing waters but an old GF-derived ^{236}U input. Unfortunately, the historical atmospheric deposition, riverine input, and oceanic transport of GF-derived ^{236}U to the Baltic Sea are not well constrained due to a lack of observation data, therefore we only made a rough estimation for different inputs of GF-derived ^{236}U .

According to the areal inventory of GF-derived ^{236}U in the Black Forest peat core (9.0×10^{12} atoms/ m^2 ; Quinto et al., 2013), the total atmospheric depositions of GF-derived ^{236}U on the Baltic Sea and its catchment were estimated to be 1.4 kg (393,000 km^2) and 5.8 kg (1630,000 km^2), respectively (Lin et al., 2021b), with the predominant deposition occurring in the 1950s–1960s (Quinto et al., 2013; Hain et al., 2020; Ohno et al., 2021). For the GF-derived ^{236}U deposited on the Baltic Sea, a maximum of 47 g of GF-derived ^{236}U directly reached the seabed (corresponding to the sedimentary inventory of GF-derived ^{236}U before 1970; Lin et al., 2021b), and the rest (1.35 kg) should be dissolved in the Baltic seawater. For the GF-derived ^{236}U deposited on the catchment, the mixing diagram suggested a low GF-derived ^{236}U concentration ($\sim 10^7$ atoms/kg) in the river runoff of the Baltic Sea, which was close to the extrapolated level in the Elbe River water to the North Sea ($< 10^7$ atoms/kg; Christl et al., 2017). Assuming an average runoff of 454 km^3 /year (Johansson, 2016), the annual riverine input of GF-derived ^{236}U to the Baltic Sea was estimated to be 1.8 g/year in 2018, which was a minor contribution to the modern Baltic seawater inventory (266 ± 5 g). For oceanic transport, a net output of GF-derived ^{236}U from the Baltic Sea to the North Sea was estimated over the past five decades (see details in Section 4.4.2). Hence, we concluded that the dissolution of deposited GF-derived ^{236}U in the Baltic seawater should be the major input of GF-derived ^{236}U .

4.4.2. Long-term dynamics of GF-derived ^{236}U

Different from the reprocessing-derived ^{236}U , there are still some knowledge gaps in the historical inputs of GF-derived ^{236}U to the Baltic Sea, therefore it was not feasible to perform a long-term hindcast simulation for GF-derived ^{236}U without reliable input functions. Instead, we used sedimentary records to uncover the temporal evolution of GF-derived ^{236}U in the Baltic Sea.

The reported GF-derived ^{236}U records resolved from coral and sediment cores suggested a general exponential decrease of GF-derived ^{236}U in global marine systems since the 1980s (Winkler et al., 2012; Sakaguchi et al., 2016; Nomura et al., 2017; Lin et al., 2021b). A comparison (Fig. 6B and C) between the simulated GF-derived ^{236}U concentrations in the surface layer of the North Sea (55°N, 5°E; data from Christl et al., 2015b) and the reconstructed GF-derived ^{236}U concentrations in the bottom waters of the Gotland Basin and the Landsort Deep (Lin et al., 2021b) revealed a longer effective half-life of GF-derived ^{236}U in the Baltic Sea (21 - 33 years) than the North Sea (11 years). This should also be related to the strong "memory effect" of the Baltic Sea. Similar to NR-derived ^{236}U , higher GF-derived ^{236}U concentrations in the Baltic brackish waters might represent the "aged" GF-derived ^{236}U signals trapped in the central Baltic Sea due to the limited water renewal.

Based on the differences between the simulated level in the North Sea and the resolved level in the Baltic Sea, we could roughly estimate the exchange of GF-derived ^{236}U between the two seas. The annual import of GF-derived ^{236}U to the Baltic Sea was estimated by multiplying the simulated GF-derived ^{236}U concentrations in the North Sea with the mean Baltic inflow (634 km^3 /year; Dahlgard, 2002). Due to the

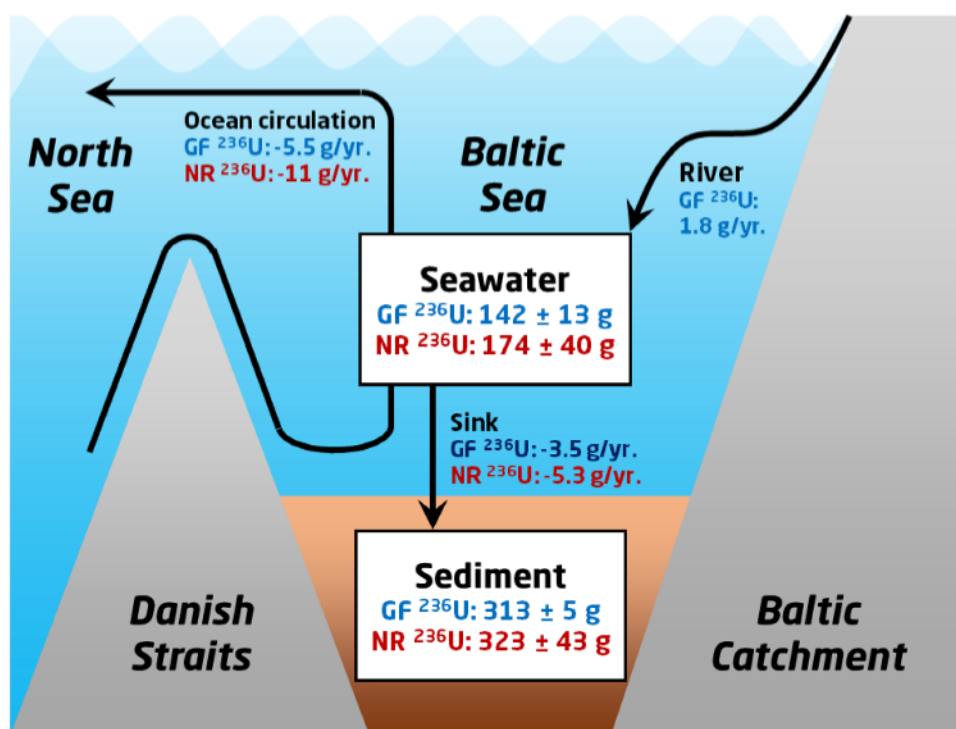


Fig. 8. General scheme of GF-derived and NR-derived ^{236}U budgets in the Baltic Sea in 2018–2019. The solid arrows represent the fluxes of GF-derived and NR-derived ^{236}U , and the positive/negative values refer to the inputs to/outputs from the Baltic seawater. The flux for the ocean circulation represents the net output from the Baltic Sea to the North Sea. The abbreviations in the plots are: GF, global fallout; NR, nuclear reactor.

homogenous distribution of GF-derived ^{236}U in the main body of the Baltic Sea (Fig. 5 and 6B) arising from its slow water circulation, we assumed that the Baltic outflowing water had the same GF-derived ^{236}U concentrations as the Baltic bottom waters. Thereby the annual export of GF-derived ^{236}U from the Baltic Sea was calculated by multiplying the resolved concentrations in the central Baltic Sea with the mean Baltic outflow ($1107 \text{ km}^3/\text{year}$; Dahlgaard, 2002). The net decadal flux of GF-derived ^{236}U between the two seas was estimated to be $73 \pm 13 \text{ g}$, $-23 \pm 15 \text{ g}$, $-63 \pm 18 \text{ g}$, $-52 \pm 2 \text{ g}$, and $-44 \pm 3 \text{ g}$ for the past five decades, respectively (positive/negative values represent input to/output from the Baltic Sea). In total, a net output of GF-derived ^{236}U ($108 \pm 41 \text{ g}$) from the Baltic Sea was obtained during 1971–2018.

4.4.3. Budget calculation for GF-derived ^{236}U

Regarding the GF-derived ^{236}U budget of the Baltic Sea during 1971–2018, we found a deficit of $((1353 - 108 - 266) - 142 =) 837 \text{ g}$ for the observed inventory of the modern Baltic seawater ($142 \pm 13 \text{ g}$) when taking all available inputs (dissolution of the atmospheric deposition, 1353 g) and outputs (oceanic transport, 108 ± 41 ; scavenging process, 266 ± 5) into account. It should be noted that this deficit could be even larger if there were significant historical riverine inputs. An over-estimated atmospheric deposition and/or an underestimated sedimentary inventory of GF-derived ^{236}U in the Baltic seawater might be the major contributors to this deficit.

In 2018–2019, the annual riverine input of GF-derived ^{236}U to the Baltic Sea was estimated to be 1.8 g (Fig. 8). The annual net output of GF-derived ^{236}U from the Baltic Sea was estimated as $5.5 \pm 0.3 \text{ g}$ based on the aforementioned calculations. The annual deposition rate of GF-derived ^{236}U to the Baltic sediments was estimated to be $3.5 \pm 1.5 \text{ g}$ (Lin et al., 2021b). In summary, the Baltic Sea lost $5.5 + 3.5 - 1.8 = 7.2 \pm 1.5 \text{ g}$ of GF-derived ^{236}U in the year of 2018. Similar to the case of NR-derived ^{236}U , the loss of GF-derived ^{236}U in the Baltic Sea water will likely continue in this decade.

5. Conclusions and perspectives

In this work, based on the large-scale observation and long-term 3D numerical simulation, we obtained in-depth knowledge on the source terms, transport, mixing, and fate of anthropogenic U in the Baltic Sea. The major findings of this work include: (1) the budget calculations for anthropogenic ^{236}U in the modern Baltic seawater indicate that the nuclear weapon testing and civil nuclear industries have comparable contributions ($142 \pm 13 \text{ g}$ and $174 \pm 40 \text{ g}$), primarily via the global atmospheric deposition in the 1950s–1960s and liquid discharges from the European reprocessing plants since the 1970s, respectively; (2) limited water renewal results in a strong "memory effect" of the Baltic Sea retaining the aged ^{236}U signals (as well as other pollutants/nutrients) for decades, and the previously unknown NR-derived ^{236}U in the Baltic Sea should be the reprocessing-derived ^{236}U discharged in the mid-1990s; (3) there is a notable sink of uranium from the water column to anoxic sediments in the Baltic Sea, which is the major contributor to the loss of ^{236}U in the Baltic seawater. Our preliminary results also demonstrate the potentials of reprocessing-derived ^{236}U and ^{129}I to trace the water-mass movement/mixing and estimate the water ages in the Baltic Sea. Combining field observations and 3D ocean modeling will enable us to further investigate the transit passages and circulation timescales of saline water in different sub-basins of the Baltic Sea, providing fundamental knowledge of the pollutant dynamics in the Baltic Sea to the decision-makers for the environmental management in the Baltic region.

Our observations reveal that uranium has unconservative behaviors in the sulfidic waters of the Baltic Sea. However, this shortcoming will not affect the robustness of using $^{233}\text{U}/^{236}\text{U}$ atomic ratios for tracing the water-mass movement and identifying emission sources of anthropogenic uranium in the Baltic Sea, as no significant isotopic fractionation occurs in the uranium scavenging processes. Besides, although the uranium depletion in the central Baltic Sea may increase the $^{129}\text{I}/\text{NR}$ -derived ^{236}U atomic ratio by up to 66% ($=100\% / (100\% - 40\%) - 100\%$) resulting in underestimated water age, considering the rapid increase

(two orders of magnitude) of the reprocessing-derived $^{129}\text{I}/^{236}\text{U}$ atomic ratios in the inflowing waters since the 1970s, the introduced uncertainties for the water-age estimation is negligible (1–3 years). Nevertheless, for a more accurate prediction of the spatiotemporal distribution of ^{236}U in the Baltic Sea, coupling sedimentation model with 3D ocean model should be considered in the future numerical simulations, where parameterization and verification for the uranium scavenging kinetics need to be investigated by laboratory experiments and field observations.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:[10.1016/j.watres.2021.117987](https://doi.org/10.1016/j.watres.2021.117987).

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