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Research Paper

Long-term environmental risks of the Baltic Sea’s "memory effect" revealed by ocean modeling and observation of reprocessing-derived radiotracers

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HIGHLIGHTS

• Baltic Sea has a strong "memory effect" retaining pollutants/nutrients for decades.
• Environmental risks of the Baltic Sea’s "memory effect" are assessed by modeling.
• 5-decade simulation is performed for radiotracer transport in the North-Baltic Sea.
• Any conservative pollutant in the Baltic Sea has ~26 yr. of environmental half-life
• The Baltic Sea annually exports 3% of contamination to the downstream areas.

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ABSTRACT

Although previous research indicated that the Baltic Sea has a strong "memory effect" for trapping pollutants/nutrients, the associated environmental risks are not well understood due to the knowledge gaps in the long-term hydrodynamics-driven exchange of pollutants/nutrients between the North Sea and the Baltic Sea. In this work, we exploited 99Tc and 129I released from the two European nuclear reprocessing plants as oceanic tracers and pollutant proxies, and performed a five-decade hindcast simulation to quantitatively estimate the fluxes and timescales of marine transport of pollutants/nutrients in the North-Baltic Sea. Modeling results underline two potential environmental risks of the Baltic Sea’s "memory effect": (1) ~26 years of environmental half-life for any existing water-soluble pollutants/nutrients in the Baltic Sea driven by its hydrodynamics; (2) the Baltic Sea as a pollutant reservoir continuously exporting 3% of contaminations per year to the downstream areas after any pollution event. Our findings provide fundamental knowledge for understanding the long-term hydrodynamics-driven pollutant/nutrient transport in the North-Baltic Sea, facilitating the future regional management of the marine environment.

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1. Introduction

The North Sea and the Baltic Sea (Fig. 1A) are connected seas located on the continental shelf of Northwest Europe with large and densely populated catchment areas and a substantial degree of industrialization, making these two seas prone to environmental pressures. Large amounts of hazardous substances and nutrients are continuously discharged into the coastal waters of both seas by land-based and marine-based anthropogenic activities, inducing serious contamination and eutrophication (HELCOM, 2018a; OSPAR, 2021a). Oceanic circulation, to a great extent, determines the fate of pollutants/nutrients (especially the water-soluble ones) in the North-Baltic Sea (HELCOM, 2018b; Svendsen et al., 2018), as oceanic-transport-derived flux and cross-basin-wide redistribution strongly affect the budgets of nutrients, heavy metals, and carbon (Larsen and Pheiffer, 1986; Vermaat et al., 2008; Kulinski and Pempkowiak, 2011; Savchuk, 2018). For instance, the North Sea has a strong counter-clockwise circulation driven by tidal currents, wind forcing, and density/pressure gradients (Otto et al., 1990; Winther and Johannessen, 2006; Sünderman and Pohlmann, 2011; Nauw et al., 2015), leading to a fast redistribution of terrestrial pollutants/nutrients entering the North Sea in the south and a cyclonic-circulation-related transport around the North Sea. Whereas the semi-enclosed Baltic Sea is only connected with the North Sea by the narrow and shallow Danish Straits and is characterized by a highly-stratified water structure and slow water renewal (Lepparanta and Myrberg, 2009), resulting in strong retention of pollutants/nutrients (HELCOM, 2018b). Owing to their diverse hydrodynamic features (especially water residence time from years in the North Sea to decades in the Baltic Sea), ecological status, and governance regimes (Ducrotoy and Elliott, 2008), the North Sea and the Baltic Sea feature very different pollutant/nutrient dynamics and thus are usually investigated separately but not as an integrated marine system. In this case, the systematic investigation of the long-term hydrodynamic exchange of pollutants/nutrients between these two seas is scarce.

However, our recent study on the anthropogenic uranium in the Baltic Sea indicated that the Baltic Sea has a strong ‘memory effect’...
trapping the aged radioactive pollutants from the North Sea for decades and gradually feeding them back to the adjacent North Sea areas afterward (M. Lin et al., 2022). Knowledge gaps still exist in the associated environmental risks of such 'memory effect', which are arising from the lack of validated modeling tool and suitable tracer approach for the quantitative estimations of the multidecadal hydrodynamic exchange of pollutants/nutrients between the North Sea and the Baltic Sea, as well as the environmental half-life of water-soluble pollutants in the Baltic Sea. The estimations of pollutant/nutrient fluxes from discrete observations at isolated spots are often unreliable (Rodhe, 1996), especially for the highly dynamic transition zone between the North Sea and the Baltic Sea (including the Skagerrak, Kattegat, and Danish Straits). The ocean models have the methodological advantage in determining the hydrodynamic transport of soluble substances in varying timescales (Rodhe, 1996). However, most kinds of the pollutants/nutrients of common concern (e.g. nitrogen and phosphorus compounds, man-made organic chemicals, heavy metals, plastic litters, etc.) originate from diffuse sources (agriculture and leaching from waste deposits), various point sources (wastewater treatment plants), and atmospheric deposition in both seas (HELCOM, 2018a; OSPAR, 2021a), introducing additional difficulties to the source characterization and data interpretation in the numerical modeling. In addition, most of these pollutants/nutrients are not conservative in the ocean, and the ocean models have to be carefully tuned for the geochemical and biological processes of the specific pollutants/nutrients, making model parameterization and verification more complicated.

Since the 1990s, radionuclides (e.g. $^{137}$Cs, $^{99m}$Tc, $^{129}$I, $^{129}$I, $^{239}$U, etc.) released from the two European nuclear reprocessing plants at Sellafield (United Kingdoms) and La Hague (France) have been recognized as useful tracers to resolve the circulation pathways and transit times of the Atlantic waters in the subpolar regions and the Arctic Ocean (Kershaw and Baxter, 1995; Povinec et al., 2003; Orre et al., 2010; Karcher et al., 2012; Casacuberta et al., 2016; Smith et al., 2021; G. Lin et al., 2022), as well as the transfer factors of the European coastal pollutions at different locations of the North Atlantic Ocean and the Arctic Ocean (Dahlgaard, 1995). Similar achievements have also been made in the North Sea (Bois et al., 1993; Guegueniat et al., 1993; Kershaw and Baxter, 1995; McCubbin et al., 2002; Hou et al., 2007a; Christl et al., 2015a) and the Baltic Sea (Hou et al., 2000; Yi et al., 2013; Qiao et al., 2020, 2021; M. Lin et al., 2022). Compared with the conventional tracers including salinity, temperature, and chemical tracers (e.g. nutrients, dissolved organic matters, chlorofluorocarbons, sulfur hexafluoride, etc.), these reprocessing-derived radionuclides have extremely low natural backgrounds, non-steady-state distributions, point-source releases, and accessible discharge histories. Owning to the long half-lives, high water solubility, and conservative behaviors in the ocean, $^{99m}$Tc ($T_{1/2} = 0.211$ Myr) and $^{129}$I ($T_{1/2} = 15.7$ Myr.) are promising oceanic tracers and pollutant proxies for the investigation of the long-term pollutant dynamics in the North-Baltic Sea. Different from other chemical and radioactive tracers, the liquid discharges from Sellafield + La Hague are the dominating sources for $^{99m}$Tc ($\sim 93\%$) and $^{129}$I ($\sim 85\%$) in the environment, respectively (Müller et al., 1977; Fabryka-Martin et al., 1985; Aarkrog et al., 1986, 1988; Paul et al., 1987; Raisbeck and Yiou, 1999; United Nations, 2000; Aldahan et al., 2007; Hou et al., 2009, 2013; Reithmeier et al., 2010; Snyder et al., 2010; HELCOM, 2020a, OSPAR, 2021b; European Commission, 2022) (Table S1), therefore, the other sources/inputs (e.g. global fallout) can be ignored for simplification. The well-constrained point-source releases of $^{99m}$Tc and $^{129}$I ensure their superior advantages over other conventional tracers and radionuclides in the hindcast simulations, including reliable input functions, distinguishable dynamics in the study area, and convenient model validation with observations of $^{99m}$Tc and $^{129}$I. For instance, most of the modeling works in the Baltic Sea simulated the transport and distribution of $^{137}$Cs ($T_{1/2} = 30.2$ yr.) from the Chernobyl accident for dose assessment and model parameterization (Toscano-Jimenez and García-Tenorio, 2004; HELCOM, 2009; Monte, 2014; Periáñez et al., 2015), but $^{137}$Cs is not suitable to precisely characterize the long-term pollutant dynamics in the North-Baltic Sea due to the relatively short half-life of $^{137}$Cs, ambiguous atmospheric deposition pattern for Chernobyl-derived $^{137}$Cs, and unignorable contributions of $^{137}$Cs from the global fallout and nuclear reprocessing plants.

The previous modeling works on the $^{99m}$Tc (Breton and Salomon, 1995; Nielsen, 1995; Schönfeld, 1995; Karcher et al., 2004; Orre et al., 2007; Simonsen et al., 2017) and $^{129}$I (Alfimov et al., 2006; Orre et al., 2010; Karcher et al., 2012; Villa et al., 2015; Smith et al., 2021) mainly focused on their oceanic transport in the North Atlantic and its shelf seas, only in two of which the Baltic Sea was included in the model domain. Based on the documented discharges from Sellafield and La Hague, Nielsen (1995) used a box model to predict the levels of $^{99m}$Tc in the North Sea and the Baltic Sea during 1955 – 1995 and calibrated the mean water exchange rates by a comparison between predicted and observed $^{99m}$Tc concentrations. Villa et al. (2015) used a Lagrangian dispersion model to investigate the transport, distribution, and inventories of reprocessing-derived $^{129}$I in the North Sea and the Baltic Sea during 1966 – 2012. However, the box model can only calculate the hydrodynamic transport of radiotracers in an average scenario (Nielsen, 1995) but not under realistic meteorological conditions, whereas the Lagrangian dispersion model has the instinct disadvantage without considering sub-grid mixing (Meier, 2007), which exhibits less accuracy for the predicted radiotracer concentrations and fluxes in the regions where the number density of particles is low (e.g. the Baltic Sea) (Arnold et al., 2012). These intrinsic limitations make the box model and Lagrangian dispersion model hard to accurately estimate the realistic long-term hydrodynamic exchange of radiotracers between the North Sea and the Baltic Sea.

This work aims to depict the long-term hydrodynamics-driven pollutant/nutrient dynamics in the North-Baltic Sea through coupling numerical modeling and ocean observations of reprocessing-derived radiotracers. We performed a 47-year (1971 – 2017) hindcast simulation on the marine transport of $^{99m}$Tc and $^{129}$I in the North-Baltic Sea using a three-dimensional (3D) Eulerian ocean model HIROMB-BOOS Model (HBM). The simulation results were validated with available time-series/spatial-distribution observations of $^{99m}$Tc and $^{129}$I in the study region. The multidecadal hydrodynamic exchange fluxes of radiotracer between the North Sea and the Baltic Sea were quantified based on the variations of radiotracer inventories in both seas for a better understanding of the long-term environmental risks of the Baltic Sea’s ‘memory effect’.

2. Methods

2.1. Hydrodynamic model

The 3D ocean model HBM adopted in this work was developed by the Danish Meteorological Institute, and the model setup has been used in our previous research on the $^{239}$U in the North-Baltic Sea (M. Lin et al., 2022). HBM is an operational model with fully dynamic two-way nesting (She et al., 2007; Berg and Poulsen, 2012), which empowers it to simulate the complex configurations of coastlines, estuaries, and sea basins in the study area. HBM is well-tuned for reproducing hydrographic conditions (She et al., 2007; Golbeck et al., 2017) and transport dynamics (She and Murawski, 2018) in the North Sea and the Baltic Sea, making it suitable for the atmosphere-ocean-ice climate change simulations (Murawski et al., 2021) and long-term studies of marine pollutants. The comprehensive validations of HBM have been reported elsewhere (Gröger et al., 2022; Golbeck et al., 2017; Nielsen, 2022). There are four model domains in the adopted HBM setup (Fig. 1A and Table S2):

- **North Atlantic Model (NOAMOD)** - a 2D mesoscale model in a 6-nautical-mile horizontal resolution that calculates sea level as a
function of wind speed and air pressure and generates boundary data (external surge) for the North Sea-Baltic Sea regional model;

- **North Sea-Baltic Sea** - a 3D regional model in a 3-nautical-mile resolution, which is two-way nested with the following two high-resolution subdomains: Transition Area and Wadden Sea.

- **Transition Area** - a high-resolution (0.5-nautical-mile) model for the waters south of Skagen and west of Bornholm to simulate the water exchange between the North Sea and the Baltic Sea in the narrow Danish Straits;

- **Wadden Sea**- a high-resolution (0.5-nautical-mile) model for the German Bight.

### 2.2. Point sources

In the North Sea-Baltic Sea model domain, 99Tc and 129I were fed at two point sources according to the documented annual discharges from the two European reprocessing plants Sellafiel and La Hague (Fig. 1C-D) (HELCOM, 2020a; OSPAR, 2021b; European Commission, 2022). A constant discharge rate was assumed for each radiotracer from each reprocessing plant within a year. As Sellafiel is located out of the North Sea-Baltic Sea model domain, its corresponding point source was moved to the Pentland Firth located in the upper left corner of the domain (Fig. 1A). A fraction of 50% (Christl et al., 2015b) and a lag time of 1 year (McCubbin et al., 2002; Povinec et al., 2003) were assumed for the transport of radionuclides from Sellafiel to the Pentland Firth by the Scotland Coastal Current.

### 2.3. Hindcast simulation

The hindcast simulations present in this work were performed at the High-Performance Computing facility of the Danish Meteorological Institute. HBM was forced by UERRA atmospheric hourly reanalysis data (ECMWF, 2022a) and produced 3D datasets of hydrographical parameters (including sea level, transport currents, ocean temperature, salinity, etc.) and radiotracer concentrations at all horizontal grid points and vertical layers at certain depths (Arakawa-C grid). Daily instantaneous model output fields of radiotracer s are available. The simulations showed a gradual freshening of the Baltic Sea over the 1.5-year spin-up and 47 years simulation period, which is a consequence of a model-resolution-related reduction of the transport through the Danish Straits leading to weaker inflows of saline waters from the North Sea into the Baltic Sea. This is a problem that is shared by many coarse grid models, and model simulations with increased horizontal resolution proved to mitigate the salinity offset problem by strongly improving the strength of modeled inflows (She and Murawski, 2018). However, increasing the model resolution from the current 3-nautical-mile resolution to 0.5-nautical-mile or higher in the Baltic Sea, as recommended by She and Murawski (2018), will increase the required computation resources by at least 33 times higher than the current setup, which is not an option for the long-term hindcast, e.g. in the Baltic Sea Model Intercomparison Project (Gröger et al., 2022) and this work. As both salinity and radiotracers in the Baltic Sea originate from the North Sea, the underestimation of salinity will eventually result in an underestimation of the transport through the Danish Straits; therefore, correction factors were calculated for the bottom and coastal grid points due to the underestimation (<10%) of radiotracer inventories. It should be mentioned that there is no special tuning of HBM for any geochemical or biological processes, as 99Tc and 129I are traditionally assumed to be conservative or near-conservative tracers (Simonsen et al., 2017; Smith et al., 2021) and the negligible proportion of 99Tc and 129I can be scavenged from the water column by particulate matters or organisms.

### 2.4. Model validation with radiotracer observations

Table 1 summarized eight time-series/spatial-distribution observation datasets of 99Tc and 129I measured and compiled by the Radioecology and Tracer Studies research group at the Department of Environmental and Resource Engineering in the Technical University of Denmark, which were used to validate the long-term performance of HBM in the North-Baltic Sea. Five time-series datasets include 129I in the seaweed collected at station Utstra on the southwestern Norwegian coast and 99Tc and 129I in the seawater and seaweed collected at stations Klint and Hesselo in the southern Kattegat during 1980–2015 (Hou et al., 2000; Qiao et al., 2020). Some of the 99Tc results are firstly published and provided in Table S3. Three spatial-distribution datasets include 129I in the surface waters of the North Sea in August 2005 (Hou et al., 2007b) and 129I in a transect of the Baltic Sea from the Danish Straits to the western Gotland Basin in August 2006 and April 2007, respectively (Yi et al., 2011). The sampling stations for the seawater and seaweed samples are shown in Fig. 1E, and the analytical methods for 99Tc and 129I in seaweed and seawater samples have been detailed in our previous papers (Hou et al., 2007b, 2000; Qiao et al., 2020). To reduce the noises from the daily variability, especially in the highly dynamic transition zone, the simulated radiotracer concentrations were converted to the monthly mean values. It should be noted that for a reasonable comparison between the simulated 129I concentrations in the seawaters and the observed 129I/127I atomic ratios in seaweed samples, the simulated 129I concentrations were normalized to 129I/127I atomic ratios according to the simulated salinity and a correlation equation of 127I concentrations (C_127I, in μg/kg) and salinity (Sal., in PSU) derived from the observations on the Baltic surface waters in 2018 (C_127I = 1.347 × Sal. - 1.346). The comparison of 129I/127I atomic ratios between seawater (simulation results) and seaweed (observation data) was feasible based on the justification that the 129I/127I atomic ratios in seaweeds are comparable to those in the ambient seawater, as verified by our earlier study (Hou et al., 2000).

The average bias and correlation coefficient are the most commonly used parameters to evaluate the reliability of simulation in previous works (Breton and Salomon, 1995; Schönfeld, 1995; Simonsen et al., 2017), which were also used in this work to quantify the deviation and consistency between observation and simulation results. The average bias was calculated for each observation dataset by the difference between the mean of the predicted values by HBM and the mean of observed values. The correlation coefficient between each observation dataset and the corresponding simulation results was calculated by the Pearson product-moment correlation analysis, and the significance of the correlation was tested by a two-tail test at a confidence level of 95%.

### 2.5. Radiotracer inventories and transport fluxes

Based on the spatiotemporal distribution of simulated radiotracers, the inventory of 99Tc or 129I in the North Sea and Baltic Sea was estimated by cumulating the 99Tc or 129I in all grid cells (the products of grid cell volumes and simulated 99Tc or 129I concentrations) in the model domain of North Sea-Baltic Sea. The boundary between the North Sea and the Baltic Sea was from Skagen, Denmark to Gothenburg, Sweden (57.7° N). Considering that the North Sea and the Baltic Sea are semi-closed systems with limited inlets and outlets, the major transport fluxes of 99Tc and 129I in the North-Baltic Sea could be estimated by the changes of radiotracer inventories in both seas according to Eqs. 1–3:

\[
\begin{align*}
\text{f}_{\text{NS} \rightarrow \text{BS}}(n) - \text{f}_{\text{BS} \rightarrow \text{NS}}(n) &= D_{\text{in}}(n) + 0.5 \times D_{\text{out}}(n-12) \\
\text{f}_{\text{BS} \rightarrow \text{NS}}(n) &= I_{\text{NS}}(n) - I_{\text{BS}}(n-1) \\
\text{f}_{\text{NS} \rightarrow \text{BS}}(n) &= I_{\text{BS}}(n) + \text{f}_{\text{NS} \rightarrow \text{BS}}(n-1)
\end{align*}
\]

where \(f_{\text{NS} \rightarrow \text{BS}}(n)\), \(f_{\text{BS} \rightarrow \text{NS}}(n)\), and \(f_{\text{NS} \rightarrow \text{NS}}(n)\) represent the discharge flux...
Table 1
Summary of the observation datasets for model validation. The bias is calculated by the difference between the mean of the predicted values by HBM and the mean of observed values; the relative bias is the ratio of the bias and the mean of observed values. The correlation coefficient is calculated by the Pearson product-moment correlation analysis, and its significance has been tested with the two-tail test at a confidence level of 95%.

<table>
<thead>
<tr>
<th>No.</th>
<th>Tracer</th>
<th>Data type</th>
<th>Period</th>
<th>Sample type</th>
<th>Sample number</th>
<th>Location</th>
<th>Observation range</th>
<th>Simulation range</th>
<th>Relative bias</th>
<th>Correlation coefficient</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>$^{129}$I/127I ratio</td>
<td>time series</td>
<td>1980–1995</td>
<td>seaweed</td>
<td>16</td>
<td>Utira, Norway (59.18° N, 4.53° E)</td>
<td>1.88–18.5 ($\times 10^{-4}$)</td>
<td>0.810–13.9 ($\times 10^{-4}$)</td>
<td>-27 %</td>
<td>0.54</td>
<td>(Jhou et al., 2000)</td>
</tr>
<tr>
<td>2</td>
<td>$^{129}$I/127I ratio</td>
<td>time series</td>
<td>1986–1999</td>
<td>seaweed</td>
<td>39</td>
<td>Klint, Denmark (55.97° N, 11.58° E)</td>
<td>3.48–37.5 ($\times 10^{-4}$)</td>
<td>5.31–57.9 ($\times 10^{-4}$)</td>
<td>19 %</td>
<td>0.93</td>
<td>(Jhou et al., 2000)</td>
</tr>
<tr>
<td>3</td>
<td>$^{99}$Tc conc.</td>
<td>time series</td>
<td>1984–1993</td>
<td>surface seawater</td>
<td>102</td>
<td>Klint, Denmark (55.97° N, 11.58° E)</td>
<td>0.131–2.68 (Bq/m$^2$)</td>
<td>0.194–1.07 (Bq/m$^2$)</td>
<td>-40 %</td>
<td>0.74</td>
<td>this work</td>
</tr>
<tr>
<td>4</td>
<td>$^{99}$Tc conc.</td>
<td>time series</td>
<td>1998–2015</td>
<td>surface seawater</td>
<td>28</td>
<td>Hessel, Denmark (56.17° N, 11.78° E)</td>
<td>0.0720–1.68 (Bq/m$^2$)</td>
<td>0.0939–1.68 (Bq/m$^2$)</td>
<td>-22 %</td>
<td>0.74</td>
<td>(Qiao et al., 2020)</td>
</tr>
<tr>
<td>5</td>
<td>$^{99}$Tc conc.</td>
<td>time series</td>
<td>1998–2013</td>
<td>bottom seawater</td>
<td>27</td>
<td>Hessel, Denmark (56.17° N, 11.78° E)</td>
<td>0.169–4.90 (Bq/m$^2$)</td>
<td>0.0563–3.01 (Bq/m$^2$)</td>
<td>-11 %</td>
<td>0.66</td>
<td>(Qiao et al., 2020)</td>
</tr>
<tr>
<td>6</td>
<td>$^{129}$I conc.</td>
<td>spatial distribution</td>
<td>Aug 2005</td>
<td>surface seawater</td>
<td>20</td>
<td>North Sea (20 stations)</td>
<td>12.7–382 ($\times 10^{-4}$ at/L)</td>
<td>20.0–562 ($\times 10^{-4}$ at/L)</td>
<td>12 %</td>
<td>0.73</td>
<td>(Jhou et al., 2007)</td>
</tr>
<tr>
<td>7</td>
<td>$^{129}$I conc.</td>
<td>spatial distribution</td>
<td>Aug 2006</td>
<td>water column</td>
<td>50</td>
<td>Baltic Sea (15 stations)</td>
<td>1.69–188 ($\times 10^{-4}$ at/L)</td>
<td>4.08–120 ($\times 10^{-4}$ at/L)</td>
<td>-10 %</td>
<td>0.91</td>
<td>(Yi et al., 2011)</td>
</tr>
<tr>
<td>8</td>
<td>$^{129}$I conc.</td>
<td>spatial distribution</td>
<td>Apr 2007</td>
<td>water column</td>
<td>76</td>
<td>Baltic Sea (18 stations)</td>
<td>1.38–173 ($\times 10^{-4}$ at/L)</td>
<td>4.55–155 ($\times 10^{-4}$ at/L)</td>
<td>24 %</td>
<td>0.90</td>
<td>(Yi et al., 2011)</td>
</tr>
</tbody>
</table>

from the two reprocessing plants to the North Sea, the exchange flux between the North Sea and the Baltic Sea (positive values refer to the net export from the North Sea to the Baltic Sea, and vice versa for the negative values), and the export flux from the North Sea to the Norwegian Sea in a certain month (n, n = 1 refers to January 1971), respectively; D$_{NS}$(n) and D$_{SB}$(n-12) represent the monthly discharge from La Hague in a certain month and that from Sellafield twelve months earlier, respectively; I$_{NS}$(n), I$_{BS}$(n), and I$_{NS}$(n-1) represent the inventory in the North Sea, the inventory in the Baltic Sea in a certain month and that one month earlier, respectively.

2.6. Calculation of environmental half-life

According to the release time from the feeding points, each radiotracer was divided into two fractions: the pre-1990 discharges and post-1990 discharges (the radiotracers released before and after 1990, respectively). With the termination of pre-1990 discharges, the remaining pre-1990 $^{99}$Tc and $^{129}$I were gradually cleared out of the North Sea and the Baltic Sea since 1991 due to the water renewal of both seas. As the water renewal in both seas was relatively stable and a fixed fraction of pre-1990 radiotracers was exported from both seas each year, an exponentially decreasing trend was observed in the pre-1990 radiotracers inventories, which can be used to estimate the environmental half-life of pre-1990 radiotracers in the Baltic Sea following a method reported in our previous work (Lin et al., 2021). Specifically, exponential curve regressions were adopted to the 5-year (1991–1995) inventory records of pre-1990 radiotracers in the North Sea and to the 24-year (1994–2017) inventory record of pre-1990 radiotracers in the Baltic Sea, respectively, resulting in the following functions:

$$I_{NS}^{pre-Tc-99}(n) = \exp(-0.0755 \times n + 21.2), \quad R^2 = 0.991$$

(4)

$$I_{BS}^{pre-Tc-99}(n) = \exp(-0.0022 \times n + 0.59), \quad R^2 = 0.997$$

(5)

$$I_{NS}^{pre-I-129}(n) = \exp(-0.0827 \times n + 24.8), \quad R^2 = 0.990$$

(6)

where $I_{NS}^{pre-Tc-99}(n)$, $I_{BS}^{pre-Tc-99}(n)$, $I_{NS}^{pre-I-129}(n)$, and $I_{BS}^{pre-I-129}(n)$ refer to the inventories of pre-1990 $^{99}$Tc in the North Sea and Baltic Sea (in TBq) and inventories of pre-1990 $^{129}$I in the North Sea and Baltic Sea (in kg) in a certain month (n, n = 1 refers to January 1971), respectively. According to the above exponential regression functions, the environmental half-lives of pre-1990 $^{99}$Tc and $^{129}$I in the North Sea ($T_{1/2}^{NS}$pre-Tc-99 and $T_{1/2}^{NS}$pre-I-129, in years) and those in the Baltic Sea ($T_{1/2}^{BS}$pre-Tc-99 and $T_{1/2}^{BS}$pre-I-129, in years) could be calculated as below:

$$T_{1/2}^{NS}^{pre-Tc-99} = \ln(2)/0.0755/12 = 0.765 \pm 0.009\text{years}$$

(8)

$$T_{1/2}^{NS}^{pre-I-129} = \ln(2)/0.0022/12 = 26.6 \pm 0.1\text{years}$$

(9)

$$T_{1/2}^{BS}^{pre-I-129} = \ln(2)/0.0827/12 = 6.98 \pm 0.009\text{years}$$

(10)

$$T_{1/2}^{BS}^{pre-I-129} = \ln(2)/0.0023/12 = 25.0 \pm 0.2\text{years}$$

(11)

3. Results

3.1. Comparison between simulation and observation

The comparison between our simulation results and observation datasets is summarized in Table 1 and shown in Fig. 2 and Figs. S1–2. The mean relative biases and correlation coefficients between the simulations and observations were from –40 to 24% and from 0.54 to 0.93, respectively, which are comparable with those reported in the previous modeling works on $^{99}$Tc and $^{129}$I (Breton and Salomon, 1995; Schönfeld, 1995; Simonsen et al., 2017) (Table S4). Considering that the measurement uncertainties of $^{99}$Tc and $^{129}$I were usually at the level of 10%, the present deviations between simulation and observation results are acceptable, which can be attributed to their short-term variabilities and the uncertainties in observations from sampling and measurements, as
well as the difference between the annually constant releases of radiotracers from the reprocessing plants in our hindcast simulation and the plume discharges in realistic scenarios (Bailly du Bois et al., 2020). In addition, the simulated results of 99[Tc] and 129[I] were consistent with time-series and spatial-distribution observations, as the significance of correlations has been verified by the two-tail test at a confidence level of 95%. For instance, the 99[Tc] peaks (in the 1980 s and 2000 s) in the time-series observations of the southern Kattegat were captured by HBM (Fig. 2C-E). The increasing trend of 129[I] before 2000 (Fig. 2A-B) and the spatial distribution of 129[I] during 2005–2007 in the North-Baltic Sea were also simulated (Figs. S1–2).

In general, the comparison indicated that HBM could decipher radiotracer transport, as well as their inter-annual and inter-seasonal variations, in the study area over the validated periods. It should be noted that there are some gap periods without validation datasets (e.g. the mid-1990 s for 99[Tc] and the 2010 s for 129[I]), during which possible errors may present in the simulation results. With acceptable deviation and consistency between the simulations and observations during the validated periods, we expect comparable bias (within ± 40 %) for the simulation results during these gap periods.

3.2. Spatiotemporal distribution of radiotracers

The spatiotemporal distributions of simulated 99[Tc] and 129[I] concentrations in the North-Baltic Sea during 1971–2017 are shown in Figs. 3–4 and Movie S1–4. Despite the inter-annual and inter-seasonal variations of the current fields, the long-term mean transport of radiotracers in the North Sea over the past five decades is dominated by a counter-clockwise circulation. The radioactive contaminations from La Hague (represented by 129[I] released after the 1990 s) were transported northwards to the southern North Sea by the European Coastal Current via the English Channel. The majority of radiotracers from Sellafield’s ‘feeding point’ (represented by the peak releases of 99[Tc] in the late-1970 s and late-1990 s) were advected southwards along the eastern British coast to the southern North Sea and mixed with the discharges from La Hague there. However, a small fraction of Sellafield-derived radiotracers were delivered by the central North Sea currents eastwards to the western coast of Denmark. The European Coastal Current transported the radiotracers from the southern North Sea to the Skagerrak along the western coastline of the Netherlands, Germany, and Denmark. In the Skagerrak, the majority of radiotracers joined the Norwegian Coastal Current and flowed northwards out of the northern boundary of the North Sea-Baltic Sea model domain along the Norwegian coastline, while a minor fraction was imported to the central Baltic Sea through the Kattegat and the Danish Straits. This fraction of the radiotracers was transported with the main counter-clockwise circulation in the central Baltic Sea. At deeper layers, basin-to-basin transport was carrying radiotracers from the Arkona Basin to the eastern Gotland Basin, and further on into the Gulf of Finland. Deeper transport into the Bothnian Sea was constrained by a sill. The counterpoint of the main circulation pattern was laid near the entrance to the Gulf of Finland. From there, the main circulation turned westwards towards the western Gotland Basin and then followed the coastlines southwards towards shallower and shallower water at the entrance of the Baltic Sea. In the southern Baltic Sea and the Danish straits, the radionuclides were carried by the Baltic outflowing currents towards the Skagerrak and the Kattegat, where they were carried further by the Norwegian Coastal Current.

Due to the fast water renewal of the North Sea, the temporal evolutions of simulated 99[Tc] and 129[I] in the coastal waters of the North Sea have a quick response (within 2 years) to the discharges of radiotracers from the two reprocessing plants. The peak releases of 99[Tc] from Sellafield in the late-1970 s and late-1990 s, embedded by a smaller one from La Hague in the late-1980 s, resulted in several fast counter-clockwise redistributions of 99[Tc] in the North Sea (Fig. 3A–C and Movie S1–2). After the peak releases, the 99[Tc] concentration declined quickly in the coastal areas of the North Sea after the 2000 s (Fig. 3D). Different from 99[Tc], 129[I] has a monotonically increasing discharge trend from La Hague in the 1990 s followed by relatively constant releases. Therefore, elevated 129[I] concentrations (> 10¹¹ atoms/L) can be observed in the western European coastal waters after the 1990 s (Fig. 4C–E and Movie S3–4). Owing to the slow water renewal of the Baltic Sea, the temporal evolutions of simulated 99[Tc] and 129[I] in the Baltic Sea are much less sensitive to the discharge patterns of radiotracers than those in the North Sea. During the past four decades, a much smaller variation of radiotracer concentrations was observed in the central Baltic Sea (1 order of magnitude) than in the western European coastal waters (2 – 3 orders of magnitude).

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3.3. Inventories and fluxes of radiotracers

Based on the simulation results, the inventories of 99[Tc] and 129[I] in the North Sea and Baltic Sea were estimated, and the major fluxes in the study area were thereby quantified according to the variations of radiotracer inventories in the North Sea and the Baltic Sea to evaluate the long-term hydrodynamic transport of radiotracers.

The simulated temporal evolutions of the 99[Tc] and 129[I] inventories in the North Sea and the Baltic Sea are shown in Fig. 5, which generally follow the discharge patterns of radiotracers from the two repressing plants with a flattened shape and a lag time. Due to the short water residence time (1 – 2 years) in the North Sea, the radiotracer inventories...
in the North Sea were approximately equivalent to two-year discharges from the reprocessing plants. Whereas the radiotracer inventories in the Baltic Sea were about 1 – 2 orders of magnitudes lower than those in the North Sea, indicating that only a minor fraction of radiotracers in the European coastal waters were entering the Baltic Sea.

The estimated radiotracer fluxes in the North-Baltic Sea (Fig. 6) suggest that the prevailing radiotracer fluxes from the reprocessing plants to the North Sea \( f_{RP\rightarrow NS} \) and from the North Sea to the Norwegian Sea \( f_{NS\rightarrow NwS} \) were at least one order of magnitude higher than the episodic flux between the North Sea and Baltic Sea \( f_{NS\rightarrow BS} \). The net \( f_{RP\leftrightarrow NS} \) became further less in a longer timescale owing to the water-mass oscillation in the transition zone between the North Sea and the Baltic Sea.

Fig. 3. Distribution of simulated \(^{99}\text{Tc}\) in the North-Baltic Sea. The snapshots of simulated \(^{99}\text{Tc}\) concentration (in Bq/m\(^3\)) in the surface seawater of the North Sea and the Baltic Sea at the end of 1980 (A), 1990 (B), 2000 (C), 2010 (D), and 2017 (E), respectively. The animated hydrodynamic transport of \(^{99}\text{Tc}\) in the North-Baltic Sea during 1971–2017 is shown in Movie S1-2.
Sea, indicating that the majority (>90%) of radiotracers from the reprocessing plants flowed northwards to the Norwegian coast rather than entered the Baltic Sea.

3.4. Transit time and fractions of radiotracers

As ~92% of reprocessing-derived $^{99}$Tc were discharged from Sellafield and ~76% of reprocessing-derived $^{129}$I were discharged from La Hague (HELCOM, 2020a; OSPAR, 2021b; European Commission, 2022), $^{99}$Tc and $^{129}$I are ideal proxies to investigate the transit time and
corresponding fractions of the pollutants/nutrients from the British coastal areas to the Baltic Sea and from the western European coastal areas to the Baltic Sea, respectively.

Even though there are some arguments on the definition of "transit time" (Orre et al., 2007), here we defined it as "the time taken for the maximal effect (radiotracer inventories) from a discharge (releases of radiotracers from the reprocessing plants) to be observed at a remote site (the Baltic Sea)" following Brown et al. (2002), which is more useful for the assessment on the pollutant dynamics. In this work, the time series of simulated radiotracer inventories in the North Sea and those in the Baltic Sea were compared to obtain the best-fitted lag times between the two marine systems using the Pearson product-moment correlation analysis, which were suggested to be 30 months (2.5 years) for the transport of radiotracers from Sellafield to the Pentland Firth by the Scotland Coastal Current. Combining the best-fitted lag times and the aforementioned assumptions, the average transit times (Table S5). It should be noted that as Sellafield is located out of the model domain, its corresponding feeding point was moved to the Pentland Firth (Fig. 1A) and a fraction of 50 % (Christel et al., 2015b) and a lag of 1 year (McCubbin et al., 2002; Povinec et al., 2003) were assumed for the transport of radiotracers from Sellafield to the Pentland Firth. Combining the best-fitted lag times and the aforementioned assumptions, the average transit times were estimated to be 3.5 years for Sellafield to the Baltic Sea and 2.1 years from La Hague to the Baltic Sea, which are slightly shorter than or comparable with the previous estimations of 4 – 6 years for Sellafield discharges (Dahlgaard, 1995; Povinec et al., 2003; Nielsen et al., 2010) and 2 years for La Hague discharges (Nielsen et al., 2010), respectively.

The "spiked" discharges of $^{99}$Tc in 1979 and 1996 – 1997 and $^{129}$I during 1990 – 1999 led to the rapid increases of the radiotracer inventories in the Baltic Sea after 2 – 4 years (Fig. 5), which could be used to quantitatively estimate the fractional transport of radiotracers from the North Sea to the Baltic Sea. Taking the abovementioned transit times into account, the fractions of the reprocessing-derived $^{99}$Tc and $^{129}$I reaching the Baltic Sea ($F_{Tc-99}$ and $F_{I-129}$) were estimated by Eqs. 12 and 13 that were utilized for a box model by Nielsen et al. (2010):

$$F_{Tc-99} = \frac{I_{RS-99}(n_1 + 30)I_{NS-99}(n_0 + 30)}{I_{RP-NS-Tc-99}(n)}$$

(12)

$$F_{I-129} = \frac{I_{RS-129}(n_1 + 25)I_{NS-129}(n_0 + 25)}{I_{RP-NS-I-129}(n)}$$

(13)

where $n_0 - n_1$ represent the peak release periods of $^{99}$Tc (January 1979 - December 1979 and January 1996 - December 1997) and $^{129}$I (January 1990 - December 1999); $F_{Tc-99}$ and $F_{I-129}$ were calculated as the ratios of the changes of radiotracer inventories in the Baltic Sea ($I_{RS-99}(n_1 + 30) - I_{RS-99}(n_0 + 30)$ and $I_{RS-129}(n_1 + 25) - I_{RS-129}(n_0 + 25)$) to the total discharges of radiotracers during the peak release periods ($\int_{n_0}^{n_1} I_{RP-NS-Tc-99}(n) \, dn$ and $\int_{n_0}^{n_1} I_{RP-NS-I-129}(n) \, dn$), representing the responses of the Baltic Sea to the "spiked" reprocessing-derived discharges after 25 – 30 month transport.

The calculations revealed that about 1.3 % of La Hague-derived radiotracers and about 0.45% of Sellafield-derived radiotracers entered the Baltic Sea, respectively. Even though our estimations were subject to the uncertainties from the mixing of discharges from two reprocessing plants and the variations in meteorological conditions, they are basically in line with an earlier estimation derived from a box model by Nielsen et al. (2010) that about 1% of the reprocessing-derived radionuclides can reach the Baltic Sea.

3.5. Fate of "historical" radiotracers

Even though it is not feasible to predict the spatiotemporal...
distributions of $^{99}$Tc and $^{129}$I in the next decades without knowing their future discharges, the fate of radiotracers in the post-discharge period can still be interpreted by reviewing the behaviors of "historical" radiotracers (pre-1990 $^{99}$Tc and $^{129}$I) in the North-Baltic Sea in the following decades after leaving reprocessing plants (1991–2017 in the case of pre-1990 radiotracers).

The simulated spatiotemporal distribution of pre-1990 $^{99}$Tc and $^{129}$I and their inventories in the North Sea and the Baltic Sea are shown in Figs. 7–8 and Fig. 5, respectively. Since 1991, the termination of pre-1990 discharges induced a faster exponential decrease of pre-1990 $^{99}$Tc and $^{129}$I in the North Sea with an environmental half-life of 0.68–0.78 years (Fig. 5). By the year 2000, there were negligible amounts of pre-1990 $^{99}$Tc and $^{129}$I remaining in the North Sea (Figs. 7A and Fig. 8A) except for the Norwegian coastal areas. In contrast to the North Sea, there was a much slower decline of the pre-1990 $^{99}$Tc and $^{129}$I signals in the Baltic Sea with a much longer environmental half-life of 25–27 years. Considerable amounts of pre-1990 $^{99}$Tc and $^{129}$I could still be observed in the Baltic Sea even after 27 years (Fig. 7C and Fig. 8C), but their contributions to the total discharges were continuously decreasing with the inputs of post-1990 radiotracers and outputs of pre-1990 radiotracers (right panels in Fig. 7 and Fig. 8). Even though the pre-1990 radiotracers in the northern of Baltic Sea had much lower concentrations than those in the south, they were also harder to be cleared and replaced by the post-1990 radiotracers.

3.6. Baltic Sea’s "memory effect"

The aforementioned results suggested a limited exchange of water and accompanying substances between the North and Baltic Seas, leading to a slow response of the Baltic Sea not only in the case of accumulation of pollutants but also in the case of clearance of existing contaminations. For instance, even though the last pulse release of $^{99}$Tc from Sellafield occurred more than two decades ago, considerably high concentrations of $^{99}$Tc (about half of the maximum historical level) were still observed in the Baltic Sea in 2017. This phenomenon inspires us to use radiotracers to investigate the timescale of pollutant dynamics in the Baltic Sea.

As indicated by the temporal evolution of pre-1990 radiotracers (Fig. 5), the pollutants in the Baltic Sea has a much longer environmental half-life (~26 years) than those in the North Sea (~0.7 years), suggesting a strong "memory effect" of the Baltic Sea preserving any entered pollutants for decades. However, this "memory effect" will not become notable until a period after the pollution event. For instance, $^{99}$Tc has two pulse releases in the late-1970s and late-1990s that have almost equal discharges, and the pre-1990 $^{99}$Tc accounted for up to one-third of the total $^{99}$Tc in the Baltic Sea by 2017 (Fig. 5 and Fig. 7). In the case of $^{129}$I, due to the continuously massive discharges since the 1990s, the pre-1990 $^{129}$I fraction (2% of total $^{129}$I) was still marginal compared to the post-1990 $^{129}$I fraction in the Baltic Sea by 2017 (Fig. 5 and Fig. 8).

Besides an environmental half-life of ~26 years for any reserved pollutants, another consequence of the Baltic Sea’s "memory effect" is a long-term output of existing pollutants from the Baltic Sea to the North Sea and a subsequent transport to the Arctic Ocean through the Norwegian Coastal Current (left panels of Fig. 7 and Fig. 8). In the case of $^{99}$Tc, after cutting off the feeding of pre-1990 $^{99}$Tc, the gradual decrease of pre-1990 $^{99}$Tc inventory in the Baltic Sea after 1995, on the other hand, led to a constant output of pre-1990 $^{99}$Tc (at a rate of ~2.6% of its Baltic Sea’s inventory per year) from the Baltic Sea to the North Sea. Even though this outflow flux of pre-1990 $^{99}$Tc was negligible compared to the dominating flux from the North Sea to the Norwegian Sea (the contribution of pre-1990 $^{99}$Tc to total $^{99}$Tc was ~2% in the Norwegian Coastal Current), the Baltic Sea will become a continuous contamination source of $^{99}$Tc to the transition zone and Norwegian coastal areas for decades after the termination of reprocessing discharges.

4. Discussion

Although the behaviors of pollutants/nutrients in the aquatic ecosystems can be significantly controlled by their physicochemical and biochemical properties, the environmental consequences of the Baltic Sea’s "memory effect" arise from the different hydrodynamic features between the North Sea and the Baltic Sea, which affect the fate and transport of all water-soluble pollutants/nutrients to a certain extent.

For example, the "memory effect" of the Baltic Sea has been supported by our recent investigation of the global-fallout-derived $^{236}$U in the Baltic seawater and sediments (Lin et al., 2021; M. Lin et al., 2022). After the intensive atmospheric deposition of $^{236}$U in the 1960s, there is a general decline of global-fallout-derived $^{236}$U in the surface water of the world’s oceans. Sedimentary records and numerical modeling suggested a slower decrease of global-fallout-derived $^{236}$U in the central Baltic Sea than in the Baltic Sea during the recent four decades, leading to a net export (avg. 2–6 g/yr.) of global-fallout-derived $^{236}$U from the Baltic Sea to the North Sea since the 1980s.

A more explicit example of the "memory effect" is the serious $^{137}$Cs contamination in the Baltic Sea from the Chernobyl accident. The Chernobyl accident in 1986 significantly elevated the $^{137}$Cs inventory in the Baltic Sea by at least a factor of 10 (Povinec et al., 2003), and $^{137}$Cs concentrations in seawater have not decreased back to the pre-Chernobyl levels after four decades (HELCOM, 2018a). The outflowing Chernobyl-derived $^{137}$Cs from the Baltic Sea has been observed in the Skagerrak and along the Norwegian coast (Povinec et al., 2003). The modeling results indicated that the outflow from the North Sea has persistent contribution (even though less important than those from global fallout and nuclear reprocessing plants) to the $^{137}$Cs in the Northern North Sea and the Norwegian Sea since the 1990s (Maderich et al., 2021).

In addition, the Baltic Sea’s "memory effect" can also be verified by the slow decline of polybrominated diphenyl ethers (PBDEs) in the Baltic
Sea and the consistent export to the North Sea. PBDEs have significant bioaccumulation and biomagnification in the food chain (including zooplankton and marine animals). PBDEs are regarded as the most concerning hazardous substances in the Baltic Sea, because their concentrations in biota are exceeding the threshold level by 1–3 orders of magnitude at all monitoring stations (HELCOM, 2020b). Even though the emissions of PBDEs have decreased since 2010 and some groups of PBDEs are declining in the Baltic Sea, it still needs at least 20 – 40 years for the Baltic Sea to recover from the PBDEs contamination according to the current decline rate (HELCOM, 2020b). In contrast, PBDEs are

Fig. 7. Distribution of pre-1990 $^{99}$Tc in the North-Baltic Sea. Left panels: simulated distribution of $^{99}$Tc (in Bq/m$^3$) discharged before 1990 in the surface seawater of the North-Baltic Sea at the end of 2000 (A), 2010 (C), and 2017 (E). Right panels: distribution of percentage contribution of pre-1990 $^{99}$Tc to total $^{99}$Tc in the surface seawater of the North-Baltic Sea at the end of 2000 (B), 2010 (D), and 2017 (F).
decreasing at a much faster rate (approximately 10% per year) in the main areas of the North Sea except for the Skagerrak and Kattegat, where no significant change of PBDEs is observed (OSPAR, 2022).

Fig. 8. Distribution of pre-1990 $^{129}$I in the North-Baltic Sea. Left panels: simulated distribution of $^{129}$I (in atoms/L) discharged before 1990 in the surface seawater of the North-Baltic Sea at the end of 2000 (A), 2010 (C), and 2017 (E). Right panels: distribution of percentage contribution of pre-1990 $^{129}$I to total $^{129}$I in the surface seawater of the North-Baltic Sea at the end of 2000 (B), 2010 (D), and 2017 (F).

5. Conclusions

It is worth mentioning that the current levels of reprocessing-derived $^{99m}$Tc, $^{129}$I, and $^{238}$U would not trigger any radiological threats to the ecosystems of the North Sea and the Baltic Sea. However, our
investigation of the long-term hydrodynamic exchange of these radio-tracers between the North Sea and the Baltic Sea highlights two potential environmental risks:

a) Any preserved pollutant/nutrient in the Baltic Sea might take up to a century (4 environmental half-lives) to be cleaned to < 10% of its original contamination level through oceanic circulation;

b) Any pollution event in the North-Baltic Sea will not only induce persistent damage to the local marine ecosystems but also turn the Baltic Sea into a constant contamination source to the downstream areas for decades, including the North-Baltic Sea transition zone, the Norwegian coastal areas, and the Arctic Ocean.

Our findings provide new perceptions of the pollutant/nutrient dynamics in the North-Baltic Sea. Especially, the long-time influence of the outflowing pollutants from the Baltic Sea has not been addressed in other works, which is important for designing an effective counter-measure strategy for a sustainable marine environment in the North-Baltic Sea region and beyond.

CRediT authorship contribution statement
J.L. performed the numerical modeling and drafted the manuscript; J.S. coordinated the computing resources for hindcast simulation; J.S. and J.M. instructed the model parameterization and data analysis; X.H. provided suggestions on data interpretation; J.Q. designed and coordinated the research; all co-authors participated in the discussion, reviewing, and revision of the manuscript.

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.

Data Availability
Data will be made available on request. All data needed to evaluate the conclusions in the paper are present in the paper and Supplementary Information. Additional data related to this paper can be requested from the authors.

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Environmental Implication
The controlled point source releases of radioactive substances from the European reprocessing plants and their accessible discharge histories make the reprocessing-derived radioisotopes ideal tracers to investigate the regional hydrodynamic processes for determining the fate and transport of pollutants/nutrients in the North Sea-Baltic Sea. The slow water renewal endows the Baltic Sea with a strong “memory effect” retaining pollutants/nutrients for decades. In this work, we assessed the potential environmental risks arising from the Baltic Sea’s “memory effect” by coupling ocean modeling and observation of radiotracers, which underlines the long-term impacts of this “memory effect” on the downstream areas.

Appendix A. Supporting information
Supplementary data related to this article can be found in the online version at doi:10.1016/j.jhazmat.2022.130144.

References


