



Sediments as sinks and sources of marine radionuclides: Implications for their use as ocean tracers

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ABSTRACT

A Lagrangian transport model for the North Atlantic has been applied to simulate the historical releases of ¹³⁷Cs, ¹²⁹I and ²³⁶U from the European nuclear fuel reprocessing plants. Advection by currents, mixing and decay are included, as radionuclide interactions between water, sediments and suspended matter. The model was validated comparing predictions with measured radionuclide concentrations in water and sediments in several areas. ¹²⁹I and ²³⁶U signals entering the Arctic Ocean have been compared with the input terms: the ²³⁶U signal is distorted, but the ¹²⁹I signal preserves its shape. In the first moments after the releases, the sediments act as sinks for ²³⁶U, but not significantly for ¹²⁹I and ultimately they become sources of ²³⁶U to the open sea. This results in a weaker correlation between input and output signals for ²³⁶U than for ¹²⁹I. The same effects as for ²³⁶U have been found for ¹³⁷Cs signal into the Arctic.

1. Introduction

Radionuclide releases from the European nuclear fuel reprocessing plants (NFRPs) Sellafield, UK (SF) and La Hague, France (LH) have been used to trace water mass transport in the North Atlantic Ocean. In particular, ²³⁶U has been used in the last years as a useful and powerful tracer due to its solubility, mainly in combination with ¹²⁹I (Shao et al., 2022; Casacuberta et al., 2016, 2018; Castrillejo et al., 2017), under the assumption that both radionuclides present an essentially perfect conservative behaviour in sea water (i.e., they remain completely dissolved without any interaction with suspended matter particles and sediments). The combined use of both radionuclides allows, for example, to determine circulation timescales (Wefing et al., 2021), water mass composition (Wefing et al., 2022) and transport pathways (Wefing et al., 2019). The use of these radionuclides as oceanic tracers requires to examine their geochemical properties when they are released from the reprocessing plants in their particular chemical forms. ¹²⁹I and ¹³⁷Cs properties in seawater have been evaluated and reviewed extensively [e.g. Inomata and Aoyama, 2022; Qiao et al., 2020 for ¹³⁷Cs and Hou et al., 2009 for ¹²⁹I]. However, only recently the ²³⁶U geochemical behaviour has been addressed (Castrillejo et al., 2020; Villa et al., 2018).

A numerical Lagrangian model which simulates the transport of

¹³⁷Cs (Perriñez et al., 2016), ¹²⁹I (Villa et al., 2015) and ²³⁶U (Perriñez et al., 2018) in the North Atlantic Ocean, including shelf seas, was developed to simultaneously describe the pathways of the historical releases from SF and LH in the ocean. In Villa et al. (2015) ¹²⁹I was modeled as a conservative radionuclide in water, according to its geochemical behaviour described in the available bibliography, and the modeled results agreed well with measurements of dissolved ¹²⁹I in the Atlantic Ocean. For ¹³⁷Cs, in contrast, Perriñez et al. (2016) included in the model the interactions of ¹³⁷Cs with the particulate phase (suspended matter and bed sediments). The model successfully quantified its dissolution from the contaminated coastal sediments and its subsequent transport (Perriñez et al., 2016). It also predicted that a non-negligible amount of ¹³⁷Cs would be present in seawater, even after the drastic reduction in the releases of ¹³⁷Cs in mid 1980's.

When the model was applied to ²³⁶U in Perriñez et al. (2018), considering a perfectly conservative behaviour, i.e. interactions with sediments were neglected, the simulated concentrations in surface waters of the North Sea were overestimated in relation to ²³⁶U measurements in the area. However, the incorporation of water/sediment interactions to the model allowed to reproduce satisfactorily the measured concentrations of dissolved ²³⁶U. Furthermore, the study predicted that when discharged, a third of the released ²³⁶U would be

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initially trapped in bed sediments of the coastal areas close to the NFRPs. These radionuclides would be released back to the water column in subsequent years; thus, it was found the ^{236}U signal entering the Arctic Ocean was distorted with respect to the release signal from the NFRPs. Yet, further investigations are required to better understand the geochemical behaviour of ^{236}U in the vicinity of the nuclear reprocessing plants and to support and evaluate these findings.

The purpose of this work is to underpin the preliminary conclusions described in Perriñez et al. (2018) relative to the role played by sediments in ^{236}U transport. This will be done obtaining additional validation of the model: water/sediment interactions have been included in the ^{129}I simulations for the first time; then measured and calculated ^{129}I , ^{236}U and ^{137}Cs concentrations have been compared in sediment samples of the Celtic and Irish Seas. Finally the input of ^{129}I , ^{236}U and ^{137}Cs into the Arctic Ocean have been calculated [only ^{236}U flux was calculated in Perriñez et al., 2018], which allowed to quantify the differences for these radionuclides between the input terms from the NFRPs and the output signals to the Arctic Ocean.

The model is described briefly in Section 2.1 and sampling of sediment cores and measurement methods in Section 2.2. Results are presented and discussed in Section 3.

2. Methods

2.1. Description of the model

A Lagrangian transport model was used. It includes advection due to three-dimensional currents, three-dimensional mixing due to turbulence, radioactive decay and exchanges of radionuclides between water and sediments. Exchanges of radionuclides with suspended particulate matter (SPM) and deposition of these particles on the seabed are also described. Water/sediment and water/SPM exchanges are described using a dynamic model based on kinetic transfer coefficients; thus the partition of radionuclides between water and the solid phases may be out from equilibrium conditions. Adsorption and desorption processes are described by the corresponding kinetic rates k_1 and k_2 . Mixing, decay and radionuclide interactions between water and the solid phases (sediments and SPM) are solved using a stochastic method due to the Lagrangian nature of the model. A steady SPM distribution over the model domain was created, as SPM concentrations must be known to evaluate transfer of radionuclides to this phase (details are given in Perriñez et al., 2016). Vertical particle settling velocity is described by Stoke's law.

Water circulation was obtained from JAMSTEC¹ (Japan Agency for Marine-Earth Science and Technology) OFES² (Ocean global circulation model For the Earth Simulator) global ocean model. Monthly circulation data was used, as described in Villa et al. (2015). Results of this ocean model were compared with data in several regions of the Earth (including the North Atlantic) in Masumoto et al. (2004). The domain considered in this work expands from 50°W to 25°E in longitude and from 45.1°N to 75.1°N in latitude (Fig. 1). The model horizontal resolution is 0.1° with 54 vertical levels.

The desorption kinetic rate was fixed as $k_2 = 1.16 \times 10^{-5} \text{ s}^{-1}$ for the three radionuclides, which was obtained from some experiments described in Nyffeler et al. (1984). This value has been used in many previous published modelling works [see review in Perriñez et al., 2019b]. The adsorption rate k_1 , associated to the particle affinity of an element, is deduced from the desorption rate k_2 and the radionuclide distribution coefficient k_d for ocean waters as described, for instance, in Perriñez et al. (2016, 2018, 2019b). Essentially, the relation holding between these magnitudes is:

$$k_d = \frac{1}{m} \frac{k_1}{k_2} \quad (1)$$

where m is the sediment concentration. The k_d values have been compiled by IAEA (2004) and are presented in Table 1 for the considered elements. This way, the coefficient k_1 describes the different geochemical behaviour of each radionuclide.

Additional details of the model are included elsewhere (Perriñez et al., 2016, 2018; Villa et al., 2015 and references therein). In addition, the model was applied to simulate releases from Fukushima in the Pacific Ocean (Perriñez et al., 2015, 2019a; Abascal-Ruiz, 2021) and has also participated in model intercomparison exercises organized by the International Atomic Energy Agency (IAEA, 2019).

2.2. Sampling and measurement

Two sediment cores from the Celtic Sea (Fig. 1) were collected in 2015 during the DY030 sampling campaign in the new RRS Discovery cruise included in the Shelf Sea Biogeochemistry (SSB), supported by the Natural Environment Research Council of the United Kingdom (Thompson et al., 2017). Core A (51°13'59" N; 6°7'59" W) was sampled on May the 11th and Core I (50°36'0" N; 7°5'59" W), five days later, on May the 16th. Core A is 25 cm long and Core I length is 21 cm. Both cores (10 cm diameter) were sliced into segments of different thicknesses: 0.5 cm segments in the most superficial part (top 2 cm), 1 cm segments in the middle (2–5 cm), and 2 cm segments in the deepest layers to the bottom of the core.

Radionuclide profiles for both cores are presented in Abascal-Ruiz (2021), Abascal-Ruiz et al. (2023) and Lérica-Toro et al. (2022). ^{129}I and ^{236}U were measured by AMS using the 1 MV Tandem AMS system (HVEE) in Centro Nacional de Aceleradores (CNA), Sevilla, Spain (Gómez-Guzmán et al., 2012; Chamizo et al., 2015). ^{137}Cs measurement has proceeded in Centro de Investigación, Tecnología e Innovación of Sevilla (CITIUS) by gamma spectrometry using an HPGe detector system (Hurtado et al., 2007).

3. Results and discussion

The predicted and measured radionuclide concentrations are first presented in Section 3.1. Then, ^{129}I , ^{236}U and ^{137}Cs signals into the Arctic Ocean are evaluated and discussed in Section 3.2.

3.1. ^{129}I , ^{137}Cs and ^{236}U concentrations in European seas

The temporal frames of the simulations depend on the availability of release data from Sellafield and La Hague reprocessing plants: 1952–2011 for ^{137}Cs , 1952–2013 for ^{236}U and 1966–2012 for ^{129}I .

The ^{129}I simulated concentrations (including water/sediment interactions) are compared with the results from Villa et al. (2015), where the sediment fraction was not included. Fig. 2 shows averaged ^{129}I concentrations in the North Sea surface water obtained from the new simulation compared with previous results from Villa et al. (2015) and with observations from the North Sea waters (Michel et al., 2012; Christl et al., 2015a). When the interactions with sediments were included, ^{129}I concentration in water was only slightly lower than that obtained without sediment interactions. This result is coherent with the weak interaction of ^{129}I with sediments, since it presents the lowest k_d of the three considered radionuclides (Table 1). Both sets of calculations agree with ^{129}I measured concentrations in sea water, as shown in Fig. 2 (the uncertainties associated to measurements are larger than the differences between both simulations).

As mentioned in the introduction, in the case of ^{236}U there are significant differences (above one order of magnitude) in model results if water/sediment interactions are included or not (Perriñez et al., 2018).

Two sediment cores were collected in the Celtic Sea as explained in

¹ www.jamstec.go.jp.

² www.jamstec.go.jp/ofes/ofes.html.

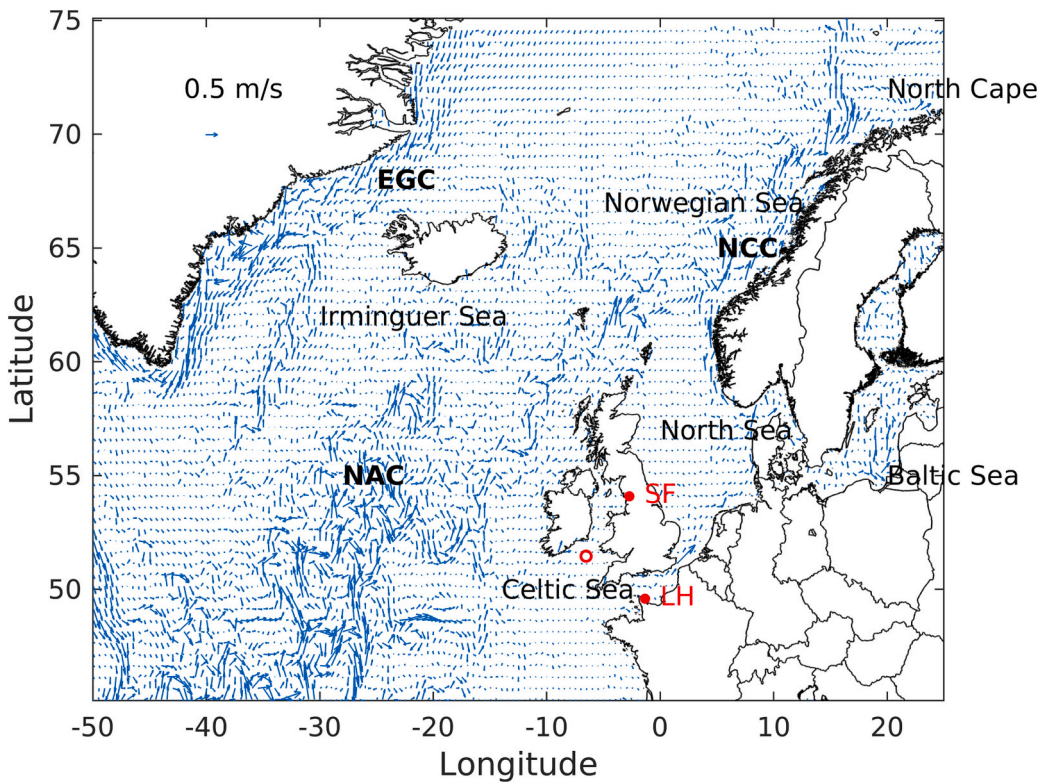


Fig. 1. Model domain. Surface circulation for January 2008 is shown as an example. Only one of each 25 vectors is drawn for more clarity. EGC: East Greenland Current, NAC: North Atlantic Current (Gulf Stream extension), NCC: Norwegian Coastal Current. SF and LH indicate Sellafield and La Hague reprocessing plants and the red open circle the position where the sediment cores mentioned in Section 2.2 were sampled. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Table 1
Dimensionless distribution coefficients in the ocean recommended by IAEA (2004) for the considered elements, which are the values used in this work.

	Cs	I	U
k_d	4×10^3	7×10^1	1×10^3

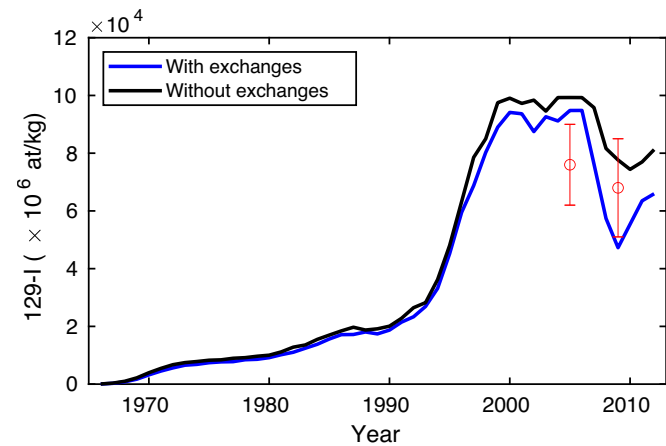


Fig. 2. Measured (points) and calculated (lines) ^{129}I concentrations in surface water of the North Sea for the simulations with and without water/sediment interactions. Measurements correspond to the calculated arithmetic mean concentrations and 1σ standard deviations over the North Sea in 2005 (from data in Michel et al., 2012) and 2009 (from data in Christl et al., 2015a).

Section 2.2. Fig. 3 compares measurements of ^{129}I , ^{137}Cs and ^{236}U in the surface layer of the cores (top 5 cm, see below) with simulation results. Water/sediment interactions were also included for ^{129}I to estimate its concentration in seabed sediments. In order to compare simulation results with measurements, the measured concentrations in the first 5 cm

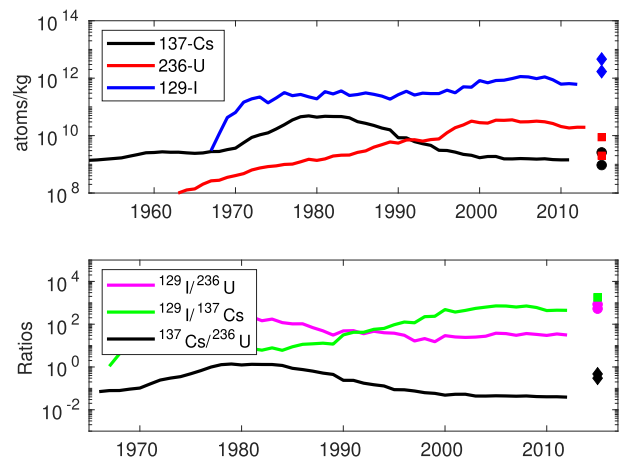


Fig. 3. Calculated (lines) and measured (points) concentrations of ^{137}Cs , ^{129}I and ^{236}U and their ratios in the two cores of the Celtic Sea. The uncertainty in measurements is within the size of the points. Only one simulation result is shown for each radionuclide because both cores are within the same grid cell of the model.

of the core were integrated, since the model considers a 5 cm thick sediment layer. Simulated and measured ratios $^{129}\text{I}/^{236}\text{U}$, $^{129}\text{I}/^{137}\text{Cs}$ and $^{137}\text{Cs}/^{236}\text{U}$ are also included in Fig. 3.

Fig. 3 shows that simulations and measurements are generally in good agreement. The simulation for ^{137}Cs provides the best results in comparison with measurements. In contrast, ^{236}U simulation results are slightly overestimated whereas ^{129}I is underestimated in the two cores. Consequently, $^{129}\text{I}/^{236}\text{U}$ ratio from the simulation is underestimated by slightly more than one order of magnitude. However, overall, the results show that the model is able to describe the adsorption of the three radionuclides by the sediments, in a position which is hundreds of km far from the radionuclide source location, SF NFRP. Note that previous

results indicate that radionuclides from LH mainly travel to the east in the English Channel, so it is assumed that most radionuclides in the Celtic Sea are coming from SF (Villa et al., 2015; Perri  nez et al., 2016, 2018).

The discrepancies in ^{129}I results might be due to the fact that the model only includes the liquid releases from Sellafield and La Hague and does not incorporate ^{129}I atmospheric emissions from the NFRPs as a secondary source to the ocean (Kadowaki et al., 2020). In Abascal-Ru  f et al. (2023) this contribution was estimated and in the Celtic Sea area 1–8 % of the ^{129}I concentration in the sediment was assigned to the atmospheric deposition from Sellafield and La Hague. This is in agreement with previous estimations obtained in similar locations (Stewart and Wilkins, 1985). Other reasons for the underestimation could be attributed to uncertainties in the k_d , since it is given in IAEA (2004) as a combination for the different iodine species, information not fully detailed in such document.

The differences between simulated and measured ^{236}U concentrations points out the complex geochemical behaviour of uranium when it is released from Sellafield NFRP. The provided k_d corresponds to a combination of two main oxidation states: U(VI), highly soluble in water, and reduced U(IV), relatively insoluble. The oxidation state of uranium when it is released from Sellafield pipeline is not known. However, uranium compounds which are insoluble (e.g. U_3O_8 , UO_2 , U metal) and partially soluble (UO_3) are produced and partially discharged (Gregory et al., 2013; IAEA, 2007) after the chemical treatment in the NFRPs. It is possible that a certain amount of ^{236}U is initially released to the coastal water in insoluble form, U(IV). This could be contributing to the overestimation of ^{236}U in the simulation.

Simulated and measured ^{129}I and ^{236}U concentrations have also been compared for seabed sediments of the Irish Sea to have an additional test of the model. Simulated concentrations were averaged over the sea, as in the case of Fig. 2 for the North Sea. Measurements have been compiled from Lee et al. (2008), Marsden et al. (2006), Pham et al. (2008) and Povinec et al. (2002). Experimental results correspond to the inventory in the first 5 cm of sediments, which correspond to approximately 7 years (Marsden et al., 2006; Morris et al., 2001). Fig. 4 shows the experimental and simulation results, where the horizontal uncertainties correspond to the seven year age of the 5 cm layer. There is a reasonable agreement between simulated and measured results.

These results confirm the role that sediments play as sink of ^{236}U in

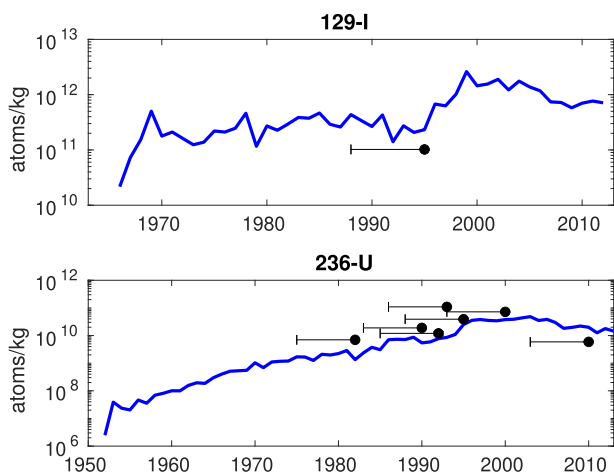


Fig. 4. Calculated (lines) and measured (points) [Lee et al., 2008; Marsden et al., 2006; Pham et al., 2008; Povinec et al., 2002] concentrations of ^{236}U and ^{129}I in bed sediments of the Irish Sea. The horizontal uncertainties correspond to the seven years age of the 5 cm layer and the uncertainties in the measured concentrations are within the size of the points. Note that the calculated concentrations correspond to averages over the Irish Sea, while measurements for specific points are provided.

the Irish Sea, in contrast to previous publications showing that the primary form of ^{236}U in open sea water is, ultimately, soluble U(VI) (Anderson et al., 1989). Christl et al. (2017) described ^{236}U conservative behaviour, i.e. in a U(VI) oxidation state, in fifty open sea water samples measured in the North sea that displayed a clear correlation between $^{236}\text{U}/^{238}\text{U}$ ratio and ^{236}U concentration. However, those results correspond to open areas of the North Sea and little is known about the geochemistry of ^{236}U in the transect from the pipelines (both SF and LH releases travel to the North Sea) to these more open waters. In any case, care should be taken when using such type of relationship between any arbitrary ratio (x/u) and x since spurious correlations result, as clearly pointed out by Stenstr  m-Khalili and H  kansson (2009), for instance.

Anderson et al. (1989) states that, despite its un-reactive behaviour in the open ocean, uranium might be removed from seawater to sediments of anoxic marine basins. There are several biogeochemical processes described by Anderson et al. (1989) which could cause the increase of uranium in the sediment phase. However, chemical reduction of soluble U(VI) to insoluble U(IV) and subsequent scavenging or interaction with organic sinking particles are not found to be a determining cause. Anderson et al. (1989) found that the main mechanism that precipitates uranium in the sediment happens when dissolved U(VI) is diffused into anoxic sediments where it is reduced and precipitated. Furthermore, note that, similarly to the coastal precipitation of releases from acid mine drainage (Villa et al., 2011), a partial co-precipitation of uranium in an insoluble compound might also occur when the acid releases from the pipeline encounter coastal seawater.

The relatively good agreement between ^{137}Cs , ^{236}U and ^{129}I measurements and simulations suggests that the model is successfully describing water/sediment interactions and that sediments play a role in retaining part of the radioactive releases of, mainly, ^{236}U and ^{137}Cs . The deviation of ^{236}U simulation results in the Celtic Sea core might be associated to small discrepancies in the estimation of the average k_d (IAEA, 2004), due to variations in the oxidation state of uranium in the coastal area before reaching the open sea and to uncertainties in its oxidation state when it is released from the NFRPs, as mentioned above. Further analysis and measurements are required to better understand the reduction process from U(VI) to U(IV) and determine its causes.

In the next section we analyze how water/sediment interactions affect the radionuclide signal out of the Irish and North Sea into the Arctic Ocean and the role of sediments not only as sink, but as a delayed source of radionuclides as well.

3.2. ^{129}I , ^{236}U and ^{137}Cs input signals into the Arctic Ocean

The fluxes of ^{129}I , ^{236}U and ^{137}Cs into the Arctic Ocean have been calculated as explained in detail in Perri  nez et al. (2018), and results are presented in Fig. 5 for ^{129}I and ^{236}U . Essentially, the calculation is done considering that the flux is $I = F \cdot C$, where C is the radionuclide concentration in the Norwegian Coastal Current and $F = 1.5 \times 10^7 \text{ m}^3/\text{s}$ is the mean flow of such current, obtained from the applied ocean model.

These fluxes are compared to the radionuclide releases from Sellafield and La Hague, which are shown in Fig. 6. ^{129}I releases were compiled from L  pez-Guti  rrez et al. (2004), Areva (2014) and Sellafield (2014). ^{236}U releases were adapted from Christl et al. (2015b), as explained in detail in Perri  nez et al. (2018). The black lines indicate the total (Sellafield plus La Hague) releases.

If a radionuclide is perfectly conservative in water, in both open and coastal waters, a correlation between the input signal (radionuclide release into the ocean) and the output signal (measured concentration somewhere downstream), is expected; with a time-delay related to the transit time between the release location and the measurement location. In our case the output is the flux to the Arctic Ocean, displayed in Fig. 5, since these fluxes are proportional to the ^{129}I and ^{236}U concentrations in the Norwegian Coastal Current, and they represent the export of the radionuclides to the Arctic Ocean, as explained above.

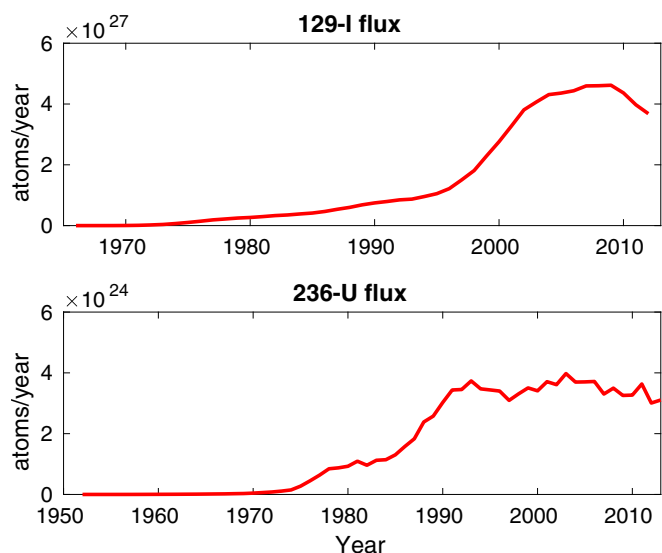


Fig. 5. Calculated fluxes of ²³⁶U and ¹²⁹I into the Arctic Ocean.

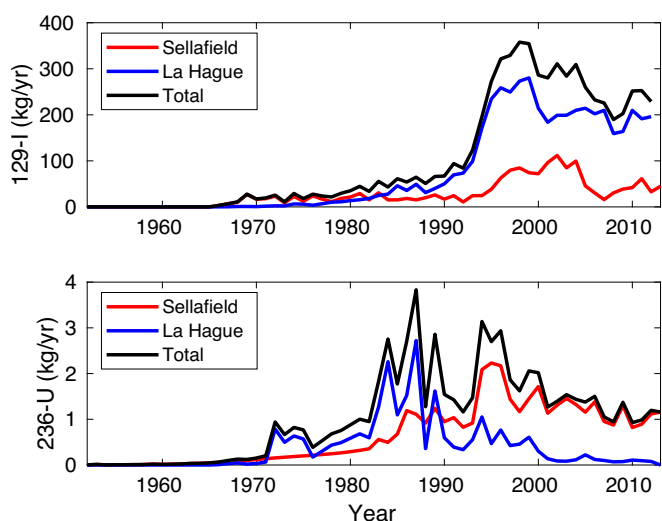


Fig. 6. Releases of ²³⁶U and ¹²⁹I from Sellafield and La Hague.

A comparison between Fig. 6 and Fig. 5 shows that the shapes of the release and export curves in the case of ¹²⁹I are fairly similar. However, this is not the case for ²³⁶U, where the peaks in releases in the 80 s and 90 s are almost completely lost, leading to a relatively smooth export flux signal. Indeed, the export signal for this radionuclide is quite similar

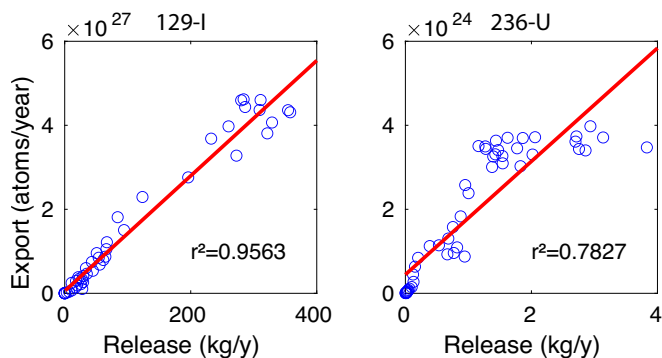


Fig. 7. Correlations between the exports to the Arctic Ocean and releases from Sellafield and La Hague.

to that of ¹²⁹I (compare both panels in Fig. 5).

This weaker correlation for ²³⁶U signals can be quantified in Fig. 7, where the best linear correlations between export signal (to the Arctic) and release signal (from NFRPs) are presented. Best correlations are obtained with delay times of 7 years and 6 years for ²³⁶U and ¹²⁹I respectively. Transit time for ²³⁶U is longer than for ¹²⁹I. This is in agreement with a less conservative behaviour of uranium than iodine in water since the mobility of a radionuclide, and hence its transit time between two locations, decreases as its k_d increases [this can be clearly seen, for instance, in the results presented in Periañez (2021, 2022)]. Finally, the more conservative behaviour of ¹²⁹I in relation to ²³⁶U can be quantified through the correlation between signals. Thus, ¹²⁹I signals present a significantly better correlation ($r^2 = 0.9563$) than the ²³⁶U signals ($r^2 = 0.7827$).

Our model is able to correctly simulate the transport of ²³⁶U and ¹²⁹I in the North Atlantic, predicting that, according to simulated and measured concentrations in water and sediments, ¹²⁹I is a more conservative tracer in both coastal and open seas. The potential sedimentation of uranium near the coast should be considered when ²³⁶U is used as a tracer of ocean circulation, since its interactions with seabed sediments cannot be totally neglected. It is important to consider, when using the ²³⁶U signal from the NFRPs into the Arctic, that the interactions with the sediment phase reduce the correlations between release and export (flux into the Arctic) signals, since ²³⁶U is deposited in sediments and later it is slowly released back to the water column. We present here evidences that sediments would behave as a long-term delayed source of ²³⁶U.

This effect is already known to occur in the case of ¹³⁷Cs in the Irish Sea after the reduction of Sellafield releases (Hunt et al., 2013; Ray et al., 2020 among many others) and also predicted for Fukushima releases in the Pacific Ocean coastal waters after the 2011 accident (Periañez et al., 2021; Periañez, 2022). We have calculated the export to the Arctic also for ¹³⁷Cs and results confirm what is outlined above. A comparison between releases from SF and LH NFRPs and the export to the Arctic for this radionuclide is presented in Fig. 8. The peak in releases (which are dominated by SF NFRP) in the 70 s is clearly detected in the flux some years later. However, although SF releases abruptly decrease in the 80 s in several orders of magnitude, this quick reduction is not so apparent in the flux, which diminishes slower than releases do. This is most likely due to the remobilisation of ¹³⁷Cs from previously contaminated sediments, now acting as a delayed radionuclide source. The best correlation between release and flux (obtained with a 8 year delay time) is presented in Fig. 9. This correlation is weaker than for ¹²⁹I and similar to that obtained for ²³⁶U (Fig. 7). Actually, the correlation for ¹³⁷Cs is

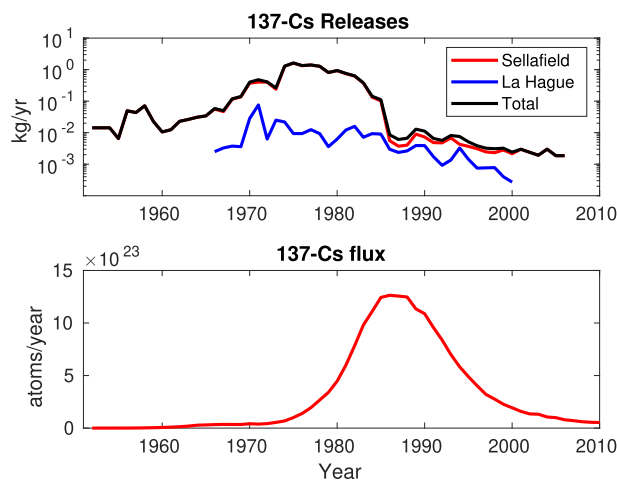


Fig. 8. ¹³⁷Cs releases from the NFRPs and calculated exports to the Arctic Ocean for this radionuclide. Releases are compiled from Areva (2014), Sellafield (2014) and Povinec et al. (2003).

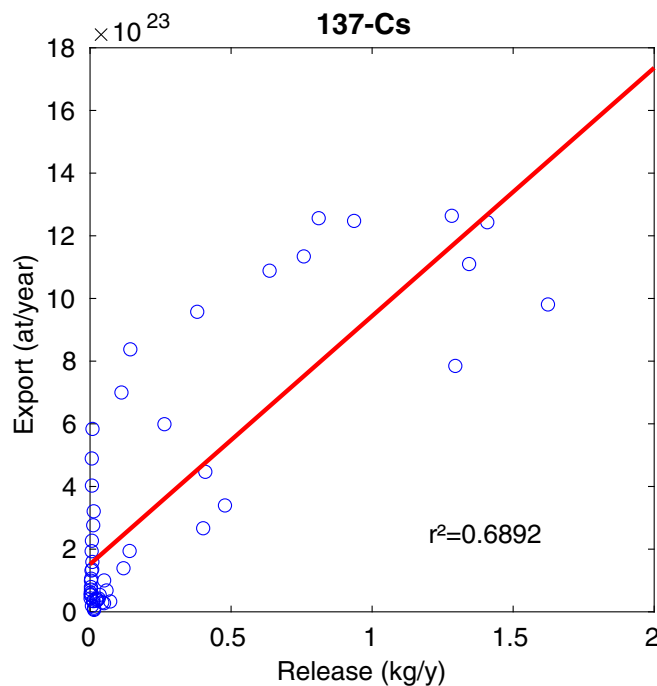


Fig. 9. Correlation between the exports to the Arctic Ocean (output signal) and releases from Sellafield and La Hague (input signal) for ^{137}Cs .

slightly worse than for ^{236}U , in agreement with the slightly higher k_d of Cs than of U (Table 1).

Sakaguchi et al. (2012) detected ^{236}U in seabed sediments of the Japan Sea at water depths larger than 2500 m. Sakaguchi et al. (2012) argue that this fact could compromise the use of this radionuclide as a conservative tracer, depending on the relative importance of ^{236}U deposition on sediments with respect to the inventory in the water column. In fact, the deposited inventory could be neglected in comparison with the dissolved inventory in these deep waters of the Japan Sea, but this may not be the general situation. Adsorption of radionuclides in sediments is higher in shallow waters (where direct adsorption from water is higher than in deep waters) and can also be in coastal waters (where suspended matter concentrations are generally larger due to river supply and coastal runoff) and thus deposition of radionuclides is enhanced (Villa et al., 2011; Marsden et al., 2006). The European Shelf Seas seem to be a clear example of these cases, where a significant fraction of the released ^{236}U (and also ^{137}Cs) is trapped in the seabed.

4. Conclusions

A Lagrangian model which simulates the transport of radionuclides in the North Atlantic has been applied to ^{137}Cs , ^{129}I and ^{236}U . The model includes advection by three dimensional currents, three dimensional turbulent mixing, radioactive decay and interactions of radionuclides between water, bed sediments and suspended matter. The last three processes are solved using stochastic techniques. Water circulation was obtained from a global ocean circulation model. The model, applied to simulate the transport of historical releases of such radionuclides from the European NFRPs, produces radionuclide concentrations generally in agreement with measurements in water and sediments.

The main conclusion of this work is that ^{236}U interactions with seabed sediments cannot be totally neglected, at least in shallow waters (e.g., the European Shelf Seas, next to the NFRPs; but this effect is likely to occur in shallow regions over the world), if this radionuclide is used as an ocean tracer. These interactions reduce the correlation between release and export signals at two locations since particle reactive radionuclides are fixed in sediments and they are later released back to the

water column. Thus, the sediments turn into an additional source of ^{236}U . The same conclusions are found in the case of ^{137}Cs , although this radionuclide has been extensively studied in the past.

Interactions with sediments are much weaker in the case of ^{129}I and consequently good release-export correlations are found; thus this radionuclide seems a suitable ocean tracer in the region under study.

CRedit authorship contribution statement

Ra  l Perri  ez: Conceptualization, Methodology, Formal analysis, Writing – original draft. **Unai Abascal-Ru  z:** Methodology, Writing – review & editing. **Jos   Maria L  pez-Guti  rrez:** Funding acquisition, Supervision. **Mar  a Villa-Alfageme:** Funding acquisition, Supervision, Methodology, Writing – review & editing.

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.

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