The behaviour of ^{236}U in the North Atlantic Ocean assessed from numerical modelling: a new evaluation of the input function into the Arctic

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Abstract

A numerical model, previously validated with other radionuclides, was applied to simulate the dispersion of ^{236}U released from European nuclear fuel reprocessing plants in the North Atlantic and Shelf Seas using a published reconstruction of Sellafield and La Hague releases. Model results are in better agreement with observations if the lowest estimation of such releases are used. This implies that approximately 40 kg of ^{236}U have been discharged from Sellafield. It was found that adsorption of ^{236}U on bed sediments of the shallow European Shelf Seas plays an essential role in its dispersion patterns. This contrasts strongly with the more

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conservative behavior of ^{129}I in the same area. This has two important implications in the use of ^{236}U as oceanographic tracer; i) special care must be taken in coastal areas, as sediments might act as sinks and sources of ^{236}U; ii) the annual input function of ^{236}U into the Arctic is not directly controlled by the annual discharges from Sellafield and La Hague, since sediments from the Irish, Celtic and North Sea modulate and smooth the signal. Only 52% of the total releases enter into the Arctic Ocean.

Keywords: Lagrangian model; North Atlantic; 236-uranium; sediments; water tracer; Nuclear Fuel Reprocessing Plants

1 Introduction

The characterization of the Arctic water masses has become fundamental in the late years, e.g., to understand how climate change would affect circulation patterns in the region. New data and updated models are of primary importance for tracing transport pathways and features of the Arctic water masses. Anthropogenic radionuclides, such as ^{137}Cs, ^{3}H, ^{99}Tc and ^{129}I, are being used as transient tracers of oceanographic processes.

^{236}U has been presented in the last years as a powerful new water mass tracer in oceanography, especially in the Atlantic Ocean and Arctic Ocean. It is almost entirely, but not exclusively, of anthropogenic origin. A total amount of 35 kg of mobile ^{236}U is estimated to come from natural sources, atmospheric nuclear weapons tests contributed to the total inventory with about 900 kg (Sakaguchi et al., 2009)). Finally, the Nuclear Fuel Reprocessing Plants (NFRP) of Sellafield, Springfields and La Hague have carried out significant liquid discharges to the sea. ^{236}U contribution from these sources has been estimated to be about 95 kg (Christl et al., 2015a). However, these estimations still need to be narrowed down. In order to use ^{236}U as a robust oceanographic tracer, it is essential

to quantify accurately the contribution of the different sources and to better define its geochemical behavior. A full three-dimensional Lagrangian model which simulates the dispersion of ^{236}U through the North Atlantic (NA) has been used to evaluate the annual discharges and the total amount of ^{236}U released in the NA from the NFRP. Furthermore, using the model it is possible to investigate, for the first time, the water/sediment interactions of ^{236}U.

The model was previously applied to study the dispersion of ^{129}I and ^{137}Cs released from the European NFRP (Villa et al., 2015; Periáñez et al., 2017, respectively) and its results were compared with measurements in both water and sediments. These are geochemically well-known radionuclides, with well-known discharges from the NFRP. It allowed us to evaluate the performances of the model. The next step has been to implement the model to improve our knowledge of ^{236}U.

The model is described in detail in references cited above and, consequently, only a brief overview is provided in section 2. Model results are presented in section 3. A reconstruction of Sellafield releases was required to run the model. Results show that the lowest estimations of such releases should be used to have a better agreement with observations. This is described in section 3.1. An investigation on ^{236}U/^{129}I ratios is presented in section 3.2, where it is shown that they must be carefully analyzed when used as water tracers, since ^{129}I is significantly more conservative than ^{236}U. Finally, a reconstruction of the ^{236}U input function into the Arctic Ocean is presented in section 3.3.

2 Model description

The model is a full three-dimensional Lagrangian dispersion model in which a radionuclide release is simulated by a number of particles, each of them equivalent to a number of units

(atoms in the present application). The three-dimensional path followed by each particle is computed, turbulent diffusion being modelled as a three-dimensional random walk process. A constant typical value of 1.0×10^{-3} m^2/s (Elliott et al., 2001) has been used for the vertical diffusivity. In a previous work on ^{129}I dispersion, it has been tested that model results are little sensitive to this parameter (Villa et al. 2015). The Smagorinsky's scheme (Cushman-Roisin and Beckers, 2011) has been adopted to describe the horizontal diffusivity.

The number of particles per water volume unit is computed to obtain radionuclide concentrations over the domain at the desired times and depths.

Interactions between the dissolved phase and solid phases (suspended matter and bed sediments) are described through a dynamic approach. Thus, uptake/release of radionuclides is considered to be described by a reversible reaction. This reaction is described by kinetic rates k_1 and k_2 as in previous works (Periáñez, 2008; 2012; Periáñez et al., 2013, among many others). A stochastic method was developed to solve the equations describing these processes. Technical details may be consulted elsewhere (Periáñez and Elliott, 2002; Periáñez and Caravaca, 2010; Villa et al., 2015; Periáñez et al., 2016). This method for describing water/sediment interactions has been used by other researches (Kobayashi et al., 2007; Min et al., 2013; Zhang and Battista, 2008) and successfully applied in model intercomparison exercises (Periáñez et al., 2015).

The considered domain in the north Atlantic extends from 50º W to 25ºE in longitude and from 45.1ºN to 75.1ºN in latitude (Fig. 1). Water circulation for the period of interest is required since currents are the main vector for the transport of tracers through advection processes. It has been obtained from JAMSTEC (Japan Agency for Marine-Earth Science and Technology) global ocean model. It is OFES (Ocean global circulation model For the Earth Simulator)[1]. A comparison of model performance with data in several regions of

[1]http://www.jamstec.go.jp/esc/research/AtmOcn/product/ofes.html♯cite_note-1

the global ocean (including the North Atlantic) may be seen in Masumoto et al., (2004). Horizontal resolution is 0.1° and there are 54 vertical levels, with increasing thickness from the surface towards the sea bottom. Monthly mean circulation has been used. As an example, surface circulation for January 2008 is shown in Fig. 1.

The model has already been successfully applied to simulate the dispersion of historical radionuclide releases from Sellafield and La Hague nuclear fuel reprocessing plant (Villa et al. 2015; Periáñez et al., 2016). In particular releases of ^{129}I, considering it as a perfectly conservative tracer, thus remaining in solution (Villa et al., 2015), and releases of ^{137}Cs (Periáñez et al., 2016). In this last case exchanges of radionuclides between water, suspended matter and bed sediments were considered. Computed and measured ^{137}Cs distributions in water and bed sediments at several areas of the European Shelf and at different times were compared. Model results were in reasonable agreement with observations (Periáñez et al., 2016).

Historical releases of ^{129}I and ^{236}U from Sellafield and La Hague for the period of interest (1952-2013, which is the time extent of the simulations) are presented in Fig. 2. In the case of ^{129}I, release data is obtained from López-Gutiérrez et al. (2004); Sellafield (2014); Areva (2014). In the case of ^{236}U information is more limited. La Hague facility has provided these releases with a few gaps. The full sequence has been reconstructed by Christl et al. (2015a). The authors have also reconstructed ^{236}U releases from Sellafield based on total uranium discharges. Three different reconstructions were carried out and the mean value and standard error were provided in Christl et al. (2015a). Such mean value is the red line in Fig. 2. Upper and lower limits, taken as the uncertainty of the mean, are indicated by the dashed black lines. The smaller releases from Springfield reprocessing plant (UK) are also given in Christl et al. (2015a) and they are included in simulations described in this paper.

3 Results

The model was initially applied to ^{129}I and compared with measurements (other than those already used in Villa et al., 2015). These are mean concentration in North Sea water in 2005 (Michel et al., 2012) and 2009 (Christl et al., 2015b); as well as a distribution map over this area in Christl et al. (2015b). Then the model was applied to ^{236}U and compared with mean concentration of this radionuclide in the North Sea in 2009 and its spatial distribution here (Christl et al., 2015b). Calculated and measured (Christl et al., 2015b) ^{129}I/^{236}U ratios were also compared. Subsequently the model was applied to evaluate the new ^{236}U input function into the Arctic.

3.1 ^{236}U and ^{129}I dispersion

Background concentrations of ^{236}U and ^{129}I due to nuclear weapon test fallout should be considered in order to compare model results and measurements. According to the estimation in Christl et al. (2015a), fallout background in the North Sea in 2009 (year and area where measurements are available), is about 10^7 at/kg for ^{236}U and about ten times smaller for ^{129}I. These uniform background values have been directly added to the calculated concentrations associated to the reprocessing plants.

Model simulations for ^{129}I, considering it as a perfectly conservative radionuclide, were described in Villa et al. (2015). To compare its behavior with the results from ^{236}U, additional results from such simulations are presented in this work. Thus, a comparison of the calculated and measured (Christl et al., 2015b) distribution in surface water of the North Sea in 2009 is presented in Fig. 3. The calculated distribution of ^{129}I is in general agreement with the measured one. A plot of measured vs. calculated concentrations can be seen in Fig. 4. Due to the Lagrangian nature of the model, not all the grid cells in which measurements are available were occupied by particles. Thus, not all observations

in Christl et al. (2015b) can be plotted. It is more adequate, in Lagrangian models, to look at general trends and averaged values over relatively larger regions than to measurements at given points. This is confirmed in the top panel of Fig. 5 where the temporal evolution of the calculated mean ^{129}I concentration over the surface water of the North Sea is shown. The calculated mean is obtained as the total ^{129}I content in the North Sea divided by the total water volume in the sea. Measured arithmetic mean concentrations, together with their 1σ standard deviations, in 2005 (Michel et al., 2012) and 2009 (Christl et al., 2015b) are also drawn. The calculated trend indicates an increase in concentrations in the beginning of the 1990s, due to the increase in La Hague releases (Fig. 2), and measured mean levels are well determined by the model. Although the model tends to overestimate measured concentrations when compared point by point (Fig. 4), general distribution patterns and mean values are in reasonable agreement with observations.

The model, that works satisfactorily in the case of ^{129}I, has been applied to ^{236}U. No calibration for this radionuclide has been carried out and initially the only change is the magnitude of the ^{236}U releases from Sellafield, La Hague and Springfield.

Calculated and measured mean concentrations in surface water of the North Sea are presented in Fig. 5 (central panel). The blue and red lines correspond, respectively, to simulations made with the mean value and the lower limit of the Sellafield releases (red line and lower black dashed lines in the bottom panel of Fig. 2). It is clear that, even with the lower estimation of the discharge, the mean ^{236}U concentration dissolved in the surface water of the North Sea in 2009 shown in the figure (75×10^6 at kg^{-1}) is overestimated by the present parameterization of the model.

The equilibrium distribution coefficient (k_d) of natural uranium, is of the same order of magnitude as that of Cs (IAEA, 2004). And since there are not indications that ^{236}U and natural uranium behave differently in the ocean (Casacuberta et al., 2016 and

157 references therein), a fraction of the released ^{236}U might be removed by the sediments
158 of the shallow shelf seas, as it actually happens for ^{137}Cs. An additional simulation was
159 carried out to test this hypothesis. Model parameters required to describe water/sediment
160 interactions are described in several references (Periáñez, 2005; Periáñez, 2008; Periáñez
161 et al., 2016). Again any model calibration has not been carried out. The model runs
162 with the same parameters as for ^{137}Cs (Periáñez et al., 2016) except the different releases
163 from the reprocessing plants and the value of the radionuclide k_d. Although the model
164 describes radionuclide exchanges between the dissolved and solid phases in a dynamic
165 way, the equilibrium k_d is used to derive the adsorption kinetic rate as explained in detail
166 in Periáñez et al. (2016) and references therein. The value recommended by IAEA (2004)
167 for coastal waters has been used for the U k_d: 1.0×10^3 (dimensionless). A summary
168 of parameters required by the model to describe water/sediment interactions is given in
169 Table 1. These parameters have been considered uniform over the whole model domain.
170 This is an approximation, due to the lack of information about their variability over such
171 a large region, which obviously will lead to some unavoidable errors in predictions. In
172 spite of this, the general behaviour of ^{236}U in sediments should be given by the model,
173 since it was successfully applied to describe patterns of ^{137}Cs in sediments with the same
174 parameter set (except the k_d).

175 The result of this new simulation is presented in the central panel of Fig. 5 (black
176 line). The "kinetics" simulation corresponds to the lower limit of Sellafield releases and
177 includes interactions of the ^{236}U between water and sediments. The mean ^{236}U dissolved
178 levels in the North Sea inferred from this new simulation is in better agreement with the
179 experimental values.

180 Additionally, the calculated inventories of ^{236}U over the model domain in bed sediments
181 in 2013 is presented in Fig. 6. It is shown that ^{236}U is mainly absorbed in the sediments of

parameter description	value
sediment mixing depth	$L = 0.05$ m
particle density	$\rho = 2600$ kg/m^3
correction factor	$\phi = 0.1$
sediment porosity	$p = 0.6$
desorption kinetic coefficient	$k_2 = 1.16 \times 10^{-5}$ s^{-1}
U distribution coefficient	$k_d = 1.0 \times 10^3$
particle radius	1.0×10^{-6} m

Table 1: Summary of model parameters involved in water/sediment interaction description. Parameter ϕ is a correction factor which takes into account that not all the sediment particle surface is available to adsorb radionuclides since may be partially hidden by other particles.

the shallow areas; this way relatively high ^{236}U concentrations are present in the Celtic Sea sediments of the continental shelf, and in the Irish Sea and English Channel (where releases from Sellafield and La Hague occur, respectively). Releases from Sellafield are higher than releases from La Hague since the 1990s (Fig. 2). Thus, inventories in the Irish Sea reach the highest values. Furthermore, releases from both plants are transported by currents to the North Sea, thus sediments in this shallow sea also show significant inventories. Values in the Celtic Sea are in the order of $10^{11} - 10^{12}$ at/m^2. The results are in agreement with the ^{236}U concentrations measured in two sediment cores collected in the area in 2015, which ranged from 10 to 20×10^{11} at/m^2 (Villa-Alfageme et al., unpublished). Calculated and measured values were higher than ^{236}U concentrations measured in sediments from the North Atlantic Porcupine Abyssal Plain (PAP) site -4500 m depth, 49º00' N, 16º30' W- (Villa-Alfageme et al. 2017). The reasons are i) ^{236}U water concentration at the PAP site is significantly lower than at the North Sea since the contribution is mostly due to fall-out ii) PAP site is a deep-sea area, where particle concentrations and downward flux are significantly lower than at the European Shelf Seas. Furthermore, modelled and measured inventories at the Celtic Sea are an order of magnitude higher than the

198 inventories measured at the Japan Sea (Sakaguchi et al., 2012), which is an area not
199 affected by NFRP discharges.

200 The calculated distribution of ^{236}U in surface water of the North Sea in 2009 is pre-
201 sented in Fig. 7 (top row), together with the map obtained after interpolation of measure-
202 ments, which has been taken from Christl et al. (2015b). Calculated levels are in relative
203 good agreement with the measured ones. However, the model largely overestimated con-
204 centrations when the medium value of the Sellafield release was used and water/sediment
205 interactions were neglected (maps not shown). A comparison of measured vs. calculated
206 concentrations in this simulation is presented in Fig. 4. The same comments as for ^{129}I
207 can be done.

208 Our results show that a) the present Sellafield releases are probably biased towards
209 the lower estimations given by Christl et al. (2015a) and b) adsorption of ^{236}U on bed
210 sediments of the shallow European Shelf Seas plays an essential role in its dispersion
211 patterns in these areas. This points out that the geochemical behaviour of ^{236}U differs
212 from that of ^{129}I in coastal areas and the continental shelf, and they might not be as
213 similar as previously predicted for open sea water (Christl et al., 2013; 2015a; 2015b).
214 While ^{129}I may be considered completely conservative, this is not the case of ^{236}U (IAEA,
215 2004). The implication of this result is that the transferences of ^{236}U between water and
216 sediments should be considered for an accurate simulation of dispersion in shallow waters.
217 Consequently, a further analysis of the ^{129}I/^{236}U ratios in coastal areas must be done when
218 this ratio is used as an complementary tracer of water mases (Casacuberta et al., 2016).

219 3.2 ^{129}I/^{236}U ratios

220 Mean calculated and measured ^{129}I/^{236}U ratios in the surface water of the North Sea in
221 2009 are presented in the bottom panel of Fig. 5. In both cases (model and measurements)

they are obtained similarly as a ratio between the mean concentrations (either from the model or measurements). The results are in good agreement within the uncertainty of the experimental values. This $^{129}I/^{236}U$ function has been also reconstructed in Christl et al. (2015a) using a single box model of the North Sea and assuming that ^{236}U is a perfectly conservative tracer (figure 10 in their paper). Such function shows a clear monotonic increase from 1990 to 2010. The present model, however, predicts quite steady values for the ratio since approximately year 2000.

Calculated and measured $^{129}I/^{236}U$ ratios, again in surface water sampled in 2009, are shown in Fig. 7 (bottom row), modelled and experimental values are in good agreement. The dark blue color in the calculation map implies background levels for the ratio. Waters coming from Sellafield and La Hague clearly show different values for the ratios, which are higher for waters entering the North Sea from the English Channel. Indeed, $^{129}I/^{236}U$ ratios in the direct releases made from Sellafield and La Hague are presented independently in Fig. 8. Ratios in discharges from La Hague are above 10^3 since 2009, while in Sellafield releases it is below 10^2.

It is noteworthy to mention that the modelled ratios calculated for both La Hague and Sellafield water entering the North Sea branches are above the ratio values from the direct discharges; this is even more apparent in the case of Sellafield, where modelled ratios are approximately an order of magnitude above those of the direct discharges. Note that the comparison with the measured data is difficult since, this Sellafield branch is essentially missed in the measurement map since there are not sampling points near the shore as required. The high ratios in relation to direct discharges are explained according to the removal of ^{236}U in bed sediments; and they are even higher in Sellafield because waters travelling from Sellafield cover a larger distance than waters coming from La Hague, thus a larger amount of ^{236}U is lost in the sediments. Consequently, the differences in the ratios

between the water branch entering the North Sea and the releases are higher in the case of Sellafield than in La Hague.

These results point out that $^{129}I/^{236}U$ ratios must be carefully analyzed when used as water tracers, since I is significantly more conservative than U (as actually in indicated by the k_d recommended for these elements by IAEA [2004]).

3.3 ^{236}U input function into the Arctic

The input function of ^{236}U into the Arctic Ocean from the Norwegian Coastal Current (NCC) has been reconstructed with the model. Thus, the annual input, I, is calculated as $I = F \cdot C$, where C is the mean concentration of ^{236}U in the NCC (which is the vector transporting material from the European Shelf into the Arctic) and F is the Norwegian Current water flow. F has been calculated from the currents provided by JAMSTEC ocean model used to drive the present dispersion model. Such flow results $F = (1.5 \pm 0.3) \times 10^7$ m^3/s at 70° N latitude (where ± 0.3 is the standard deviation from 60 monthly values). The resulting modelled ^{236}U input signal into the Arctic is given in Fig. 9, together with the signal expected from the direct releases from the different NFRP (shown in the same plot). These curves (given in kg $year^{-1}$) are integrated to obtain the total amount of ^{236}U. Thus, the total ^{236}U input released from La Hague is 24.03 kg and from Sellafield it is 40.47 kg (note that we are using the lower value of the estimation of Christl et al., 2015a). Sellafield plus La Hague input (SF+LH in Fig.9) is 64.50 kg, and Springfield releases add 8.93 kg. Finally, we calculated that the total modelled input into the Arctic (given by the integral of the black curve in Fig. 9), that is 38 ± 8 kg. This amount corresponds to only 52 ± 11 % of the total ^{236}U 73.43 kg released from Sellafield, Springfield and La Hague. The missing ^{236}U was stored in bed sediments, mostly in shallow waters, and retained in areas like the Baltic Sea, which has a mean residence time of about 10-30

years (Leppäranta and Myberg, 2009).

An interesting point is that the modelled signal into the Arctic does not reproduce the increases and decreases of the discharges of ^{236}U from Sellafield and La Hague into the ocean. There is an abrupt increase in the discharges from 1970 to approximately 1985. A secondary maximum is found in 1995 and afterwards, there is a reduction of Sellafield and La Hague ^{236}U discharges until the signal reached a rather steady value slightly lower than 1.5 kg/year. In the model, there is a monotonous increase in the input function from 1970 to approximately year 1990. After that year, the modelled input function remains constant around a value of 1.5 kg/year. Before the year 2000 the modelled signal was significantly lower than the direct discharges, this trend is reversed in 2000, were the modelled signal is remains constant, but slightly over 1.5 kg/year and the direct discharges. The reason for this evolution is that initially the sediments were acting mainly as sinks of ^{236}U, until equilibrium between the two compartments water and sediments is reached, and sediment act both as sinks and sources.

In terms of the ^{236}U signal into the Arctic, our results have two implications. First, as predicted in the previous section, the amount of ^{236}U entering into the Arctic is lower than the amount of ^{236}U directly released by the NFRP; and second, the interaction with sediment in the European Shelf shape the signal into the Arctic, and smooth abrupt changes of the direct discharges from NFRP.

4 Conclusions

A Lagrangian three-dimensional dispersion model, previously tested for ^{129}I and ^{137}Cs, has been applied to simulate the behaviour of ^{236}U released from European nuclear fuel reprocessing facilities in the North Atlantic Ocean.

The present model indicates that the actual Sellafield releases seem to be in agreement

with the lower estimations of Christl et al. (2015a). This implies a total release of about 40 kg of ^{236}U from this facility. Also, adsorption of ^{236}U on bed sediments of the shallow European Shelf Seas seem to play an essential role in its dispersion patterns in these areas. Consequently, part of the released ^{236}U is trapped in sediments. . This figure implies a total mass of 38 ± 8 kg of ^{236}U into the Arctic from the three NFRP. The input function has also been reconstructed.

It has been found that the geochemical behaviour of ^{129}I and ^{236}U is not as similar as stated in other previous works. In the first case, adsorption in sediments can be neglected, but it has to be considered for a proper dispersion simulation in the case of ^{236}U. Thus, special care should be taken when ratios between these isotopes are used to trace waters in shallow seas.

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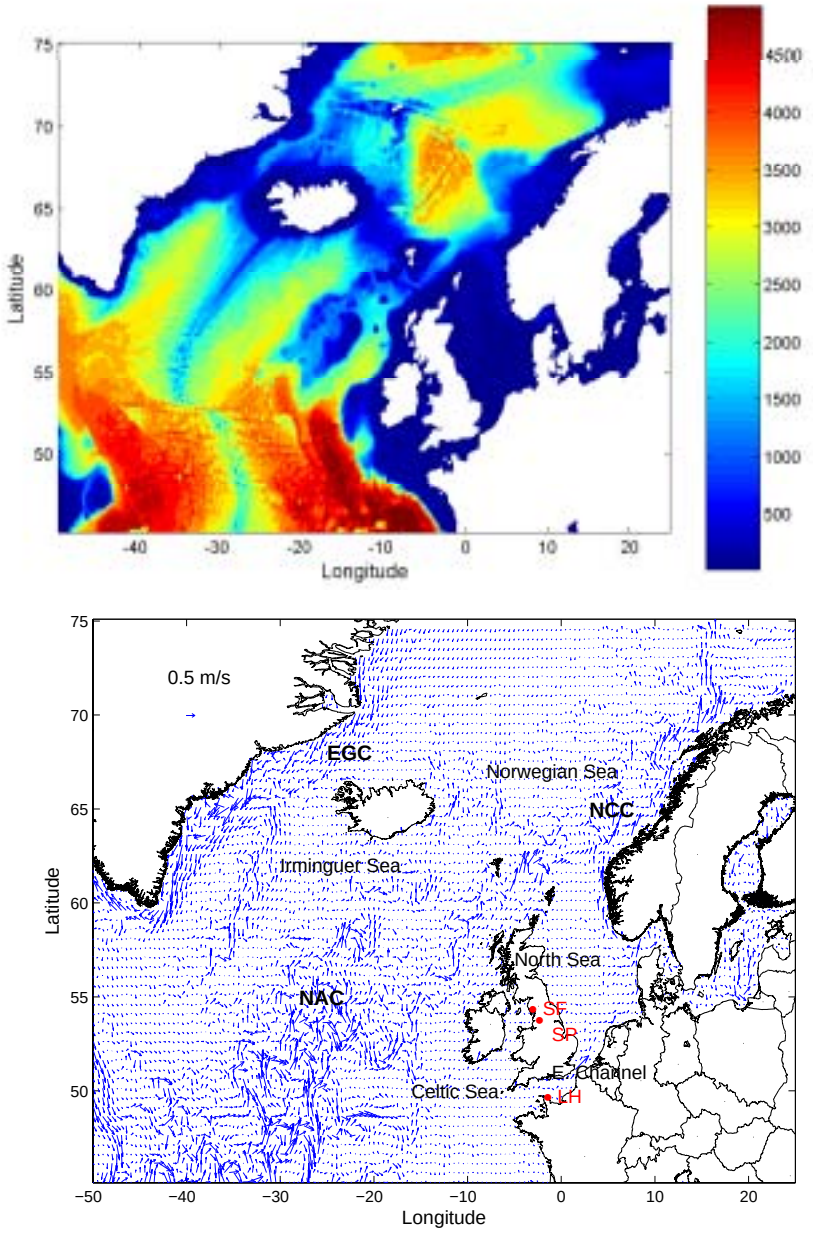

Figure 1: Top: Model domain, water depths in m. Bottom: Surface currents calculated by JAMSTEC model for January 2008 as an example. The main currents are the East Greeland Current (EGC), North Atlantic Current (NAC) and the Norwegian Coastal Current (NCC). Only one of each 25 vectors is drawn. LH, SF and SP denote La Hague, Sellafield and Springfields reprocessing plants.

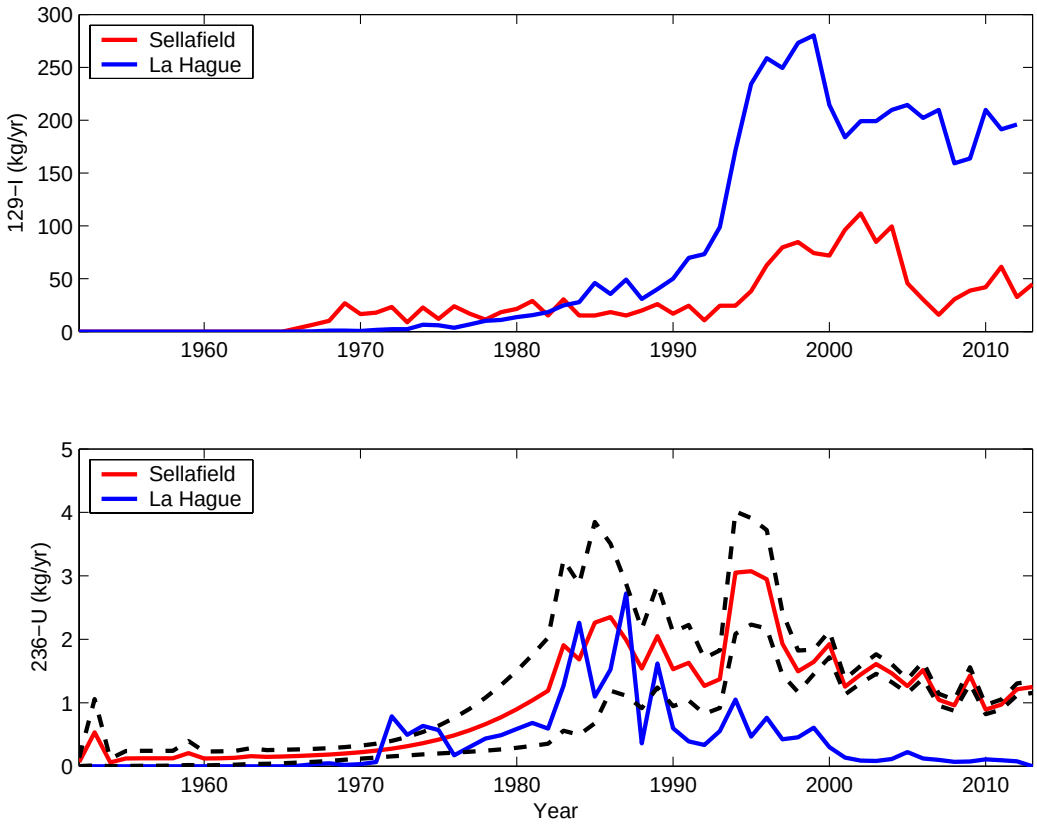

Figure 2: Annual releases of ^{129}I and ^{236}U from Sellafield and La Hague reprocessing plants. Dashed black lines indicate the lower and upper limits of the estimation of ^{236}U releases from Sellafield.

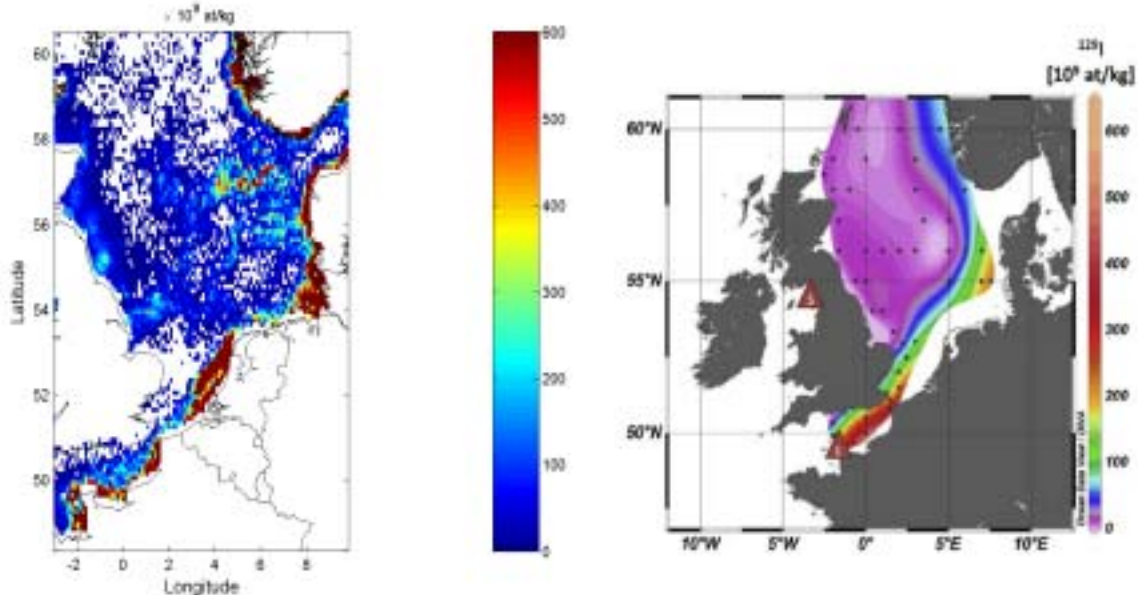

Figure 3: Calculated, left side, and measured, right side, (Christl et al., 2015b) ^{129}I concentrations in surface water of the North Sea in 2009. White areas in the sea in the calculated map indicate regions with fallout background concentration.

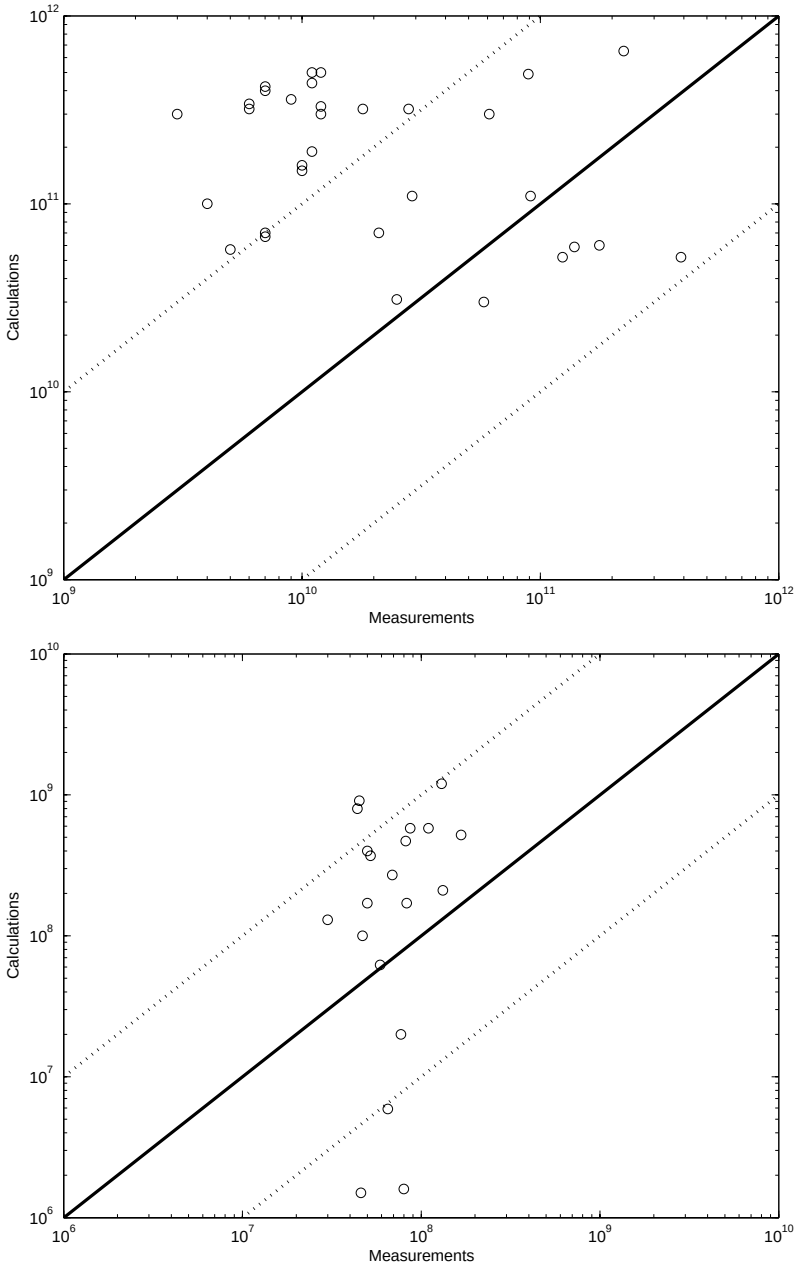

Figure 4: Calculated vs. measured concentrations (at/kg) in Christl et al. (2015b). Top and bottom panels: ^{129}I and ^{236}U respectively.

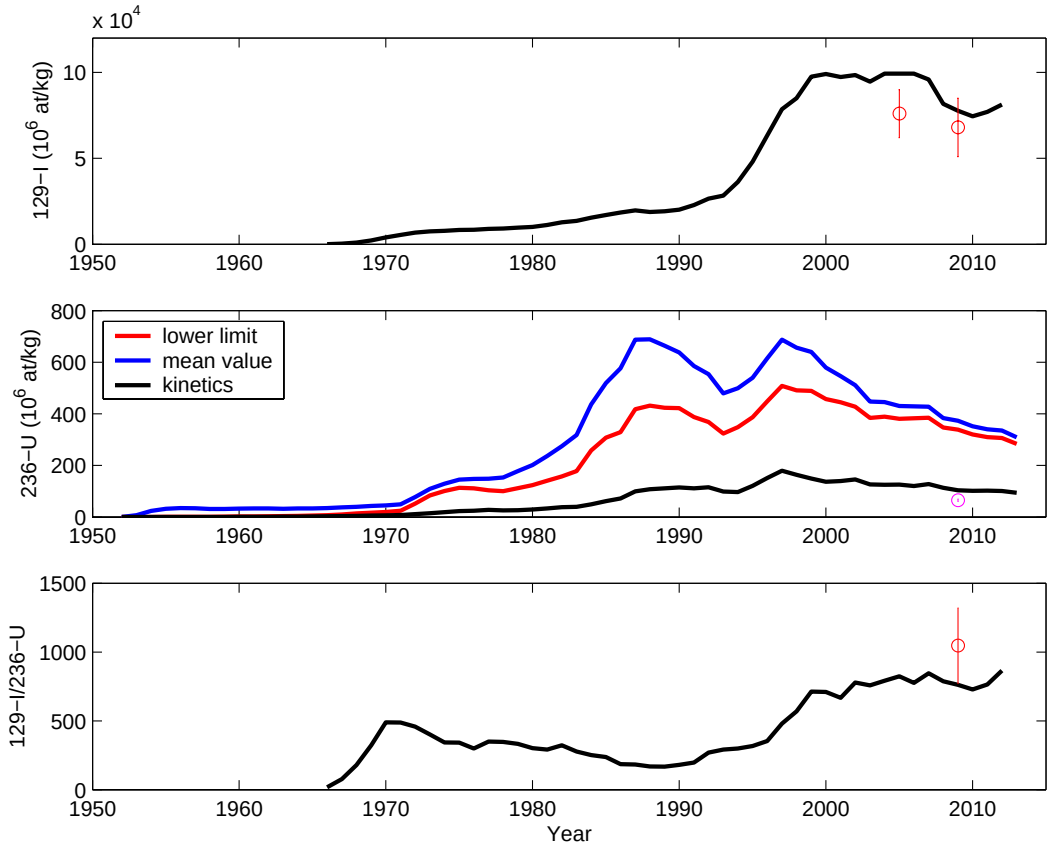

Figure 5: Top: calculated and measured mean ^{129}I concentrations in surface water of the North Sea. Middle panel: the same but for ^{236}U. See the text for explanation of the legend. Bottom: Calculated and measured mean ^{129}I/^{236}U ratios in surface water of the North Sea.

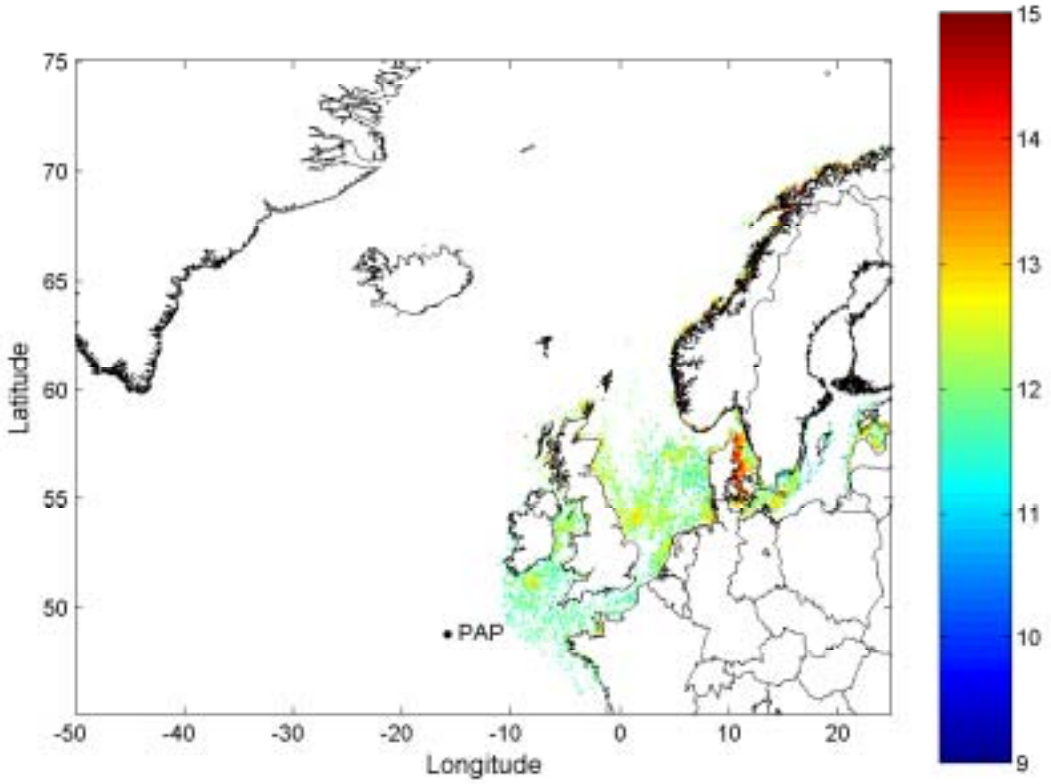

Figure 6: Calculated ^{236}U inventory (at/m^2, logarithmic scale) in bed sediments of the model domain in 2013.

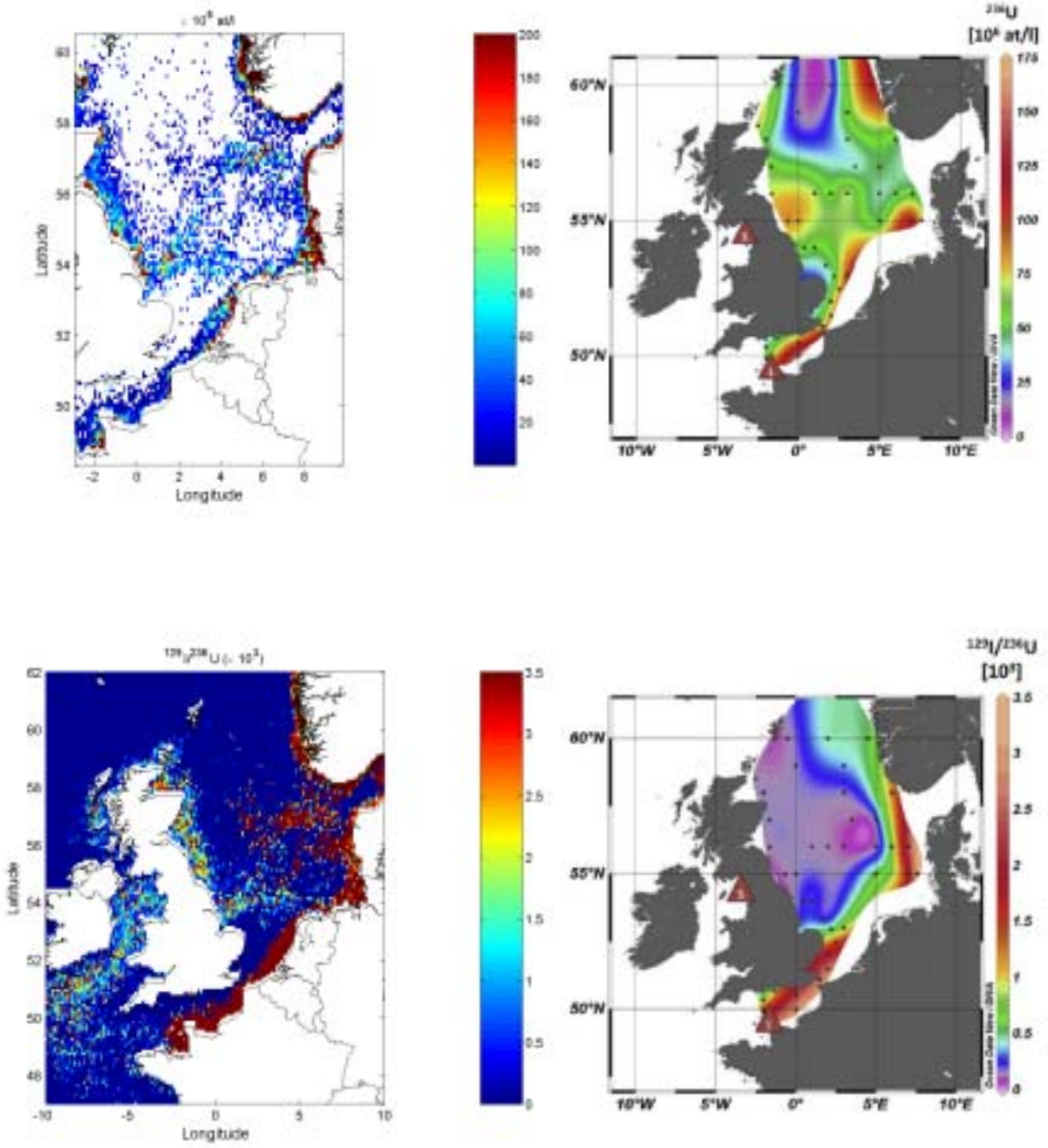

Figure 7: Calculated (left side) and measured (Christl et al., 2015b; right side) ^{236}U concentrations in surface water of the North Sea in 2009 (top row). White areas in the sea in the calculated map indicate regions with fallout background concentration. ^{129}I/^{236}U ratio in surface water (bottom row).

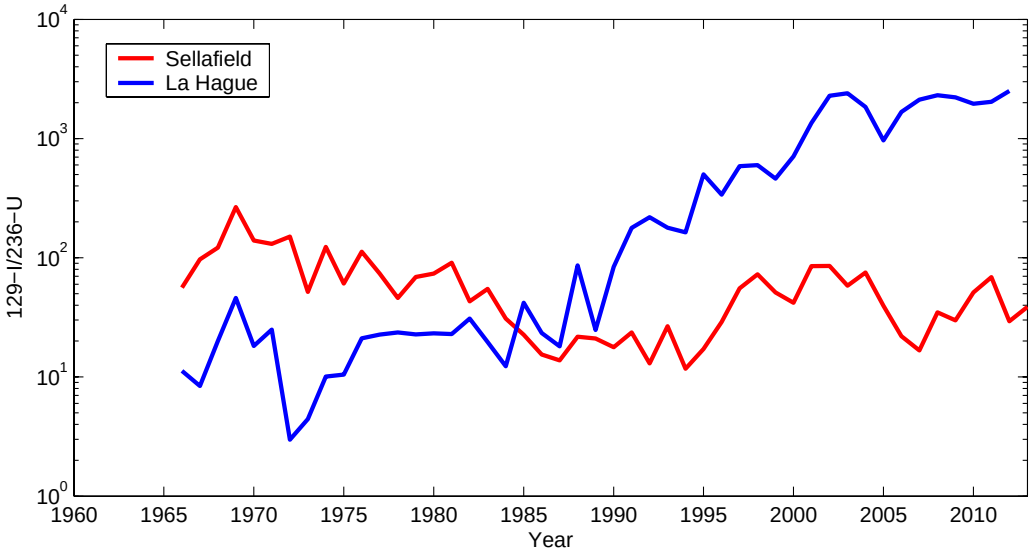

Figure 8: Annual $^{129}I/^{236}U$ ratios in the releases from Sellafield (lower value of the estimation, which is used in the model) and La Hague reprocessing plants.

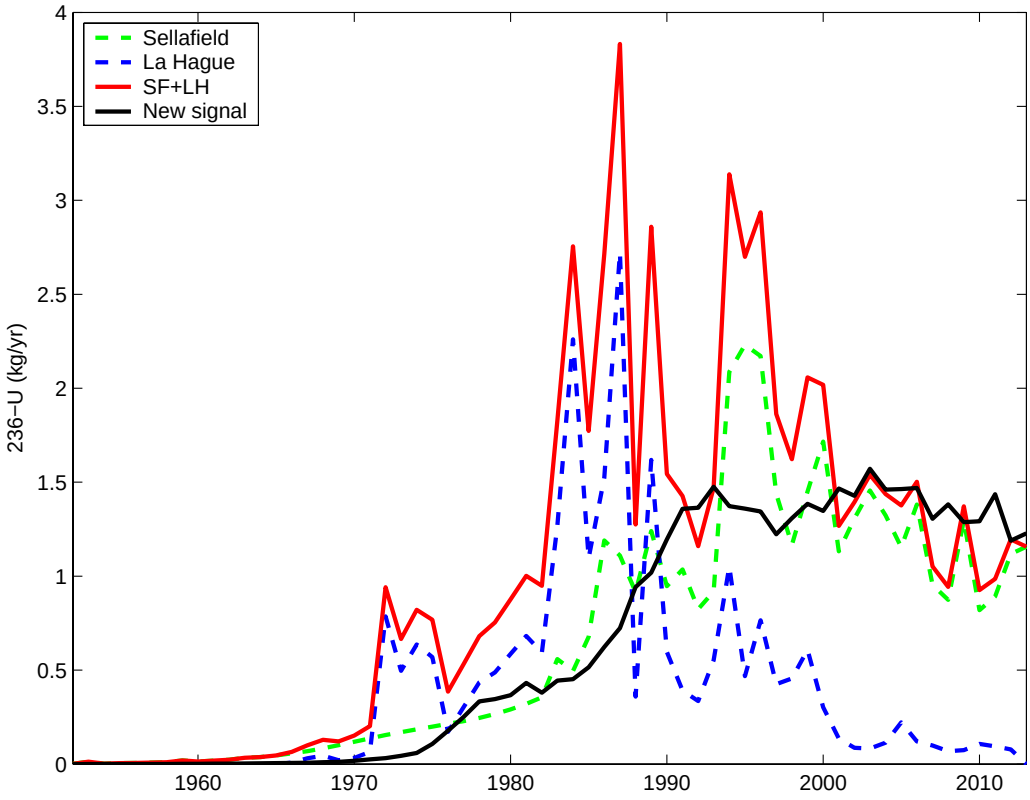

Figure 9: Annual ^{236}U input into the Arctic (black line) together with the releases from Sellafield (green dashed line), La Hague (blue dashed line) and the sum of both (red solid line).