The behaviour of ^{129}I released from nuclear fuel reprocessing factories in the North Atlantic Ocean and transport to the Arctic assessed from numerical modelling

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A B S T R A C T

A quantitative evaluation of the fate of ^{129}I, released from the European reprocessing plants of Sellafield (UK) and La Hague (France), has been made by means of a Lagrangian dispersion model. Transport of radionuclides to the Arctic Ocean has been determined. Thus, 5.1 and 16.6 TBq of ^{129}I have been intro-duced in the Arctic from Sellafield and La Hague respectively from 1966 to 2012. These figures represent, respectively, 48% and 55% of the cumulative discharge to that time. Inventories in the North Atlantic, including shelf seas, are 4.4 and 13.8 TBq coming from Sellafield and La Hague respectively. These figures are significantly different from previous estimations based on field data. The distribution of these inven-tories among several shelf seas and regions has been evaluated as well. Mean ages of tracers have been finally obtained, making use of the age-averaging hypothesis. It has been found that mean ages for Sellafield releases are about 3.5 year larger than for La Hague releases.

Keywords:
Atlantic Ocean
Iodine-129
Dispersion
Lagrangian model
Arctic Ocean

1. Introduction

Numerical models which simulate the dispersion of radionuclides in the marine environment have been continuously developed, since the pioneering works of Prandle (1984) to the most sophisticated approaches used, for instance, to predict the transport of Fukushima releases in the Pacific Ocean (Masumoto et al., 2012; Periáñez et al., 2014). Some models have also been developed to simulate the transport of radionuclides released from the European nuclear fuel reprocessing plants of Sellafield (UK) and La Hague (France) in Atlantic waters. These models have been applied to a number of radionuclides like ^{137}Cs (Prandle, 1984; Gao et al., 2004), ^{90}Sr (Gao et al., 2004), ^{99}Tc (Karcher et al., 2004) and ^{129}I (Orre et al., 2009). We are not mentioning here local-scale applications, for instance in the English Channel and in the Irish Sea.

Among these radionuclides, ^{129}I is a very significant one since, due to its biophilic behaviour and very long half-life (15.7×10^6 years), can even enter the food chain and remain there much longer than other short-lived isotopes. Thus, a number of studies concerning the distribution and fate of ^{129}I releases from the European reprocessing plants in the North Atlantic have been published (Smith et al., 2011; He et al., 2013; Michel et al., 2012; Alfimov et al., 2004, 2013; Gómez-Guzman et al., 2013).

Some models have simulated the dispersion of ^{129}I in the northern Atlantic Ocean (Orre et al., 2009), as commented above. However, only the transport pathways by the main current systems have been described. Furthermore, a quantitative estimation of the ^{129}I inventories in different sub-basins and shelf seas has never been performed. The objective of this paper consists of presenting such detailed calculations by means of a Lagrangian dispersion model. Due to the Lagrangian nature of the model, it is possible to know if a given particle, located anywhere in the domain, is coming from Sellafield or from La Hague. Thus, this allows to evaluate independently the fate of radionuclides released from each nuclear facility and the contribution of each plant to the inventories in the North Atlantic. Also, the input to the Arctic Ocean has been evaluated, quantitatively determining the fractions of Sellafield and La Hague releases which enter into this ocean. Mean age of water tracers have been calculated for releases from Sellafield, La Hague and considering both simultaneously. The age-averaging hypothesis, described below, has been adopted. The model is briefly described in Section 2. Results are presented and discussed in Section 3.

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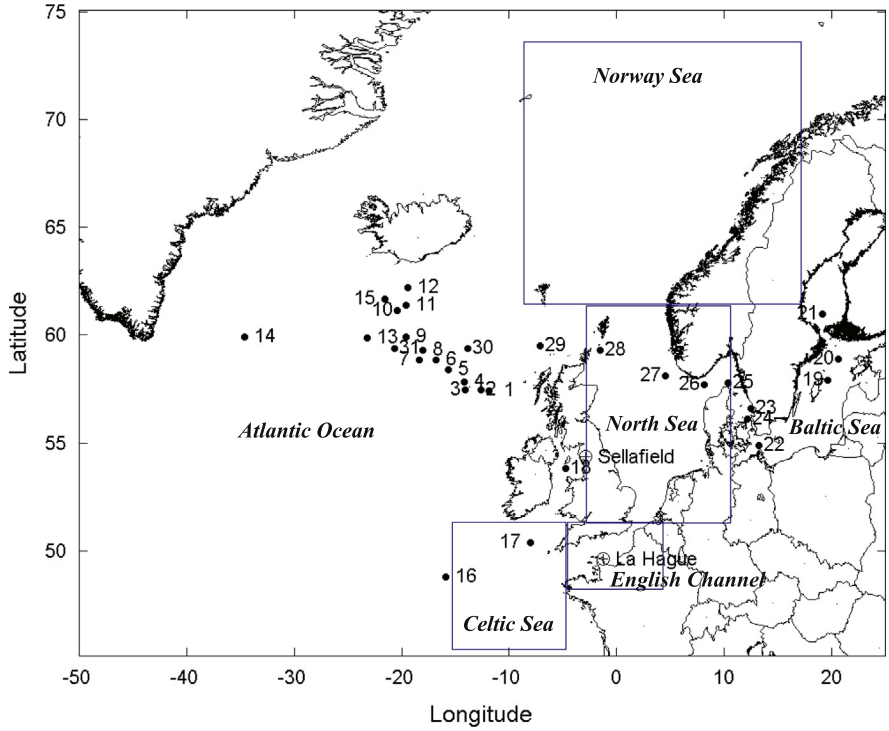

Fig. 1. Model domain with location of sampling points where measured ^{129}I concentrations have been compared with model results. Blue boxes define some regions where radionuclide contents have been evaluated. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

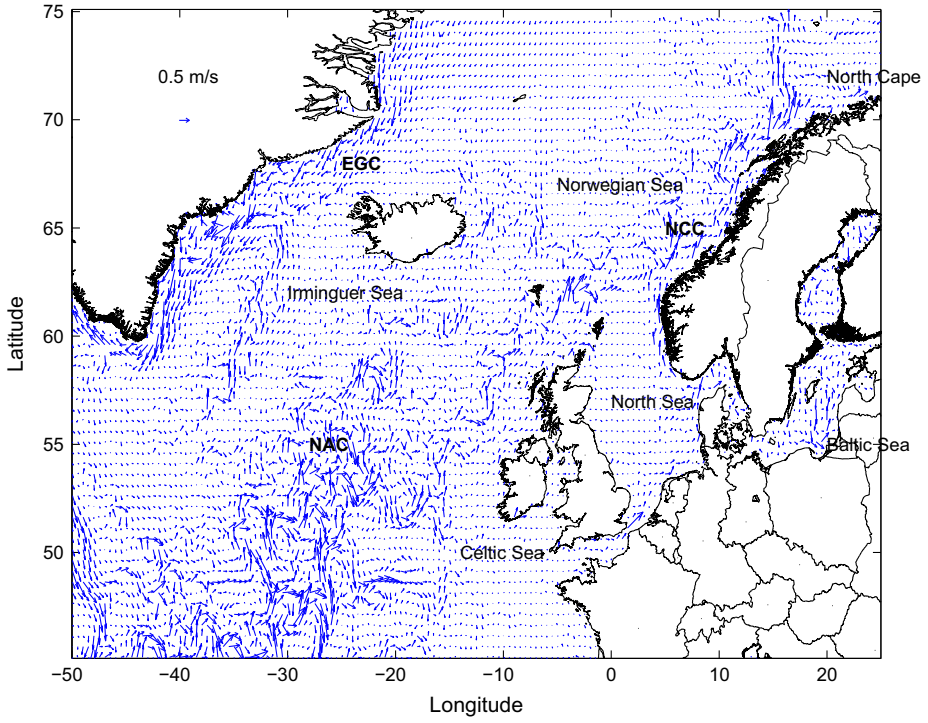

Fig. 2. Surface currents calculated by JAMSTEC model for January 2008 as an example. The main currents are the East Greenland Current (EGC), North Atlantic Current (NAC) and the Norwegian Coastal Current (NCC). Only one of each 25 vectors is drawn.

2. Model description

The model is a 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 or Bq). Lagrangian models present two advantages over Eulerian models, in which an advection/diffusion equation for mass concentration is solved. First, artificial numerical diffusion is not introduced; second, specific properties may be assigned to each particle. For instance, a "clock" may be attached to each particle, which is useful to obtain water tracer ages (Miró et al., 2012).

The three-dimensional path followed by each particle is computed, turbulent diffusion being modelled as a three-dimensional random walk process. The density of particles per water volume

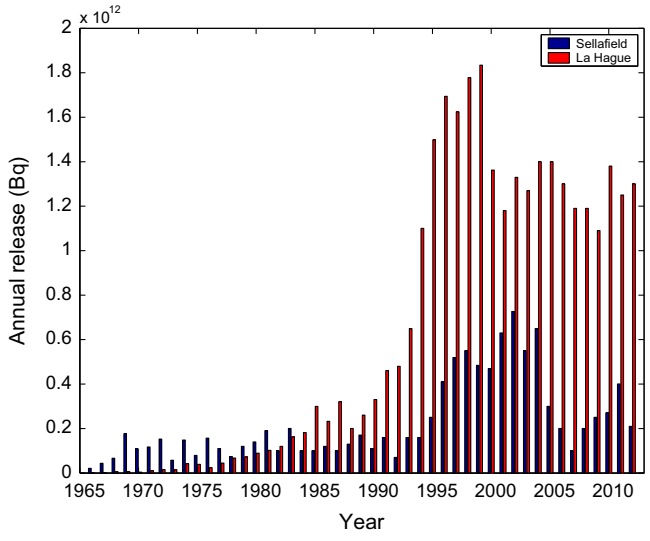

Fig. 3. Annual ^{129}I releases from Sellafield and La Hague reprocessing plants. This information has been compiled from López-Gutiérrez et al. (2004), Sellafield (2014), Areva (2014).

unit is computed to obtain radionuclide concentrations over the domain at the desired times and depths. Technical details may be consulted elsewhere (Periáñez and Elliott, 2002; Periáñez and Caravaca, 2010), but some indications are given below.

Advection is computed solving the following equation for each particle:

$$\frac{d\mathbf{r}}{dt} = \mathbf{q} \tag{1}$$

where $\mathbf{r}$ is the position vector of the particle and $\mathbf{q}$ is the current vector at the particle position and depth, solved in components u and v. Transport due to vertical advection is masked by vertical mixing due to turbulence, since vertical currents in the ocean are small. Thus, it is a common approach to neglect vertical advection in marine dispersion modelling.

The maximum size of the horizontal step given by the particle due to turbulence, D_h, is (Proctor et al., 1994; Hunter, 1987; Periáñez and Elliott, 2002):

$$D_h = \sqrt{12 K_h \Delta t} \tag{2}$$

in the direction $\theta = 2\pi RAN$, where RAN is a random number between 0 and 1. This equation gives the maximum size of the step.

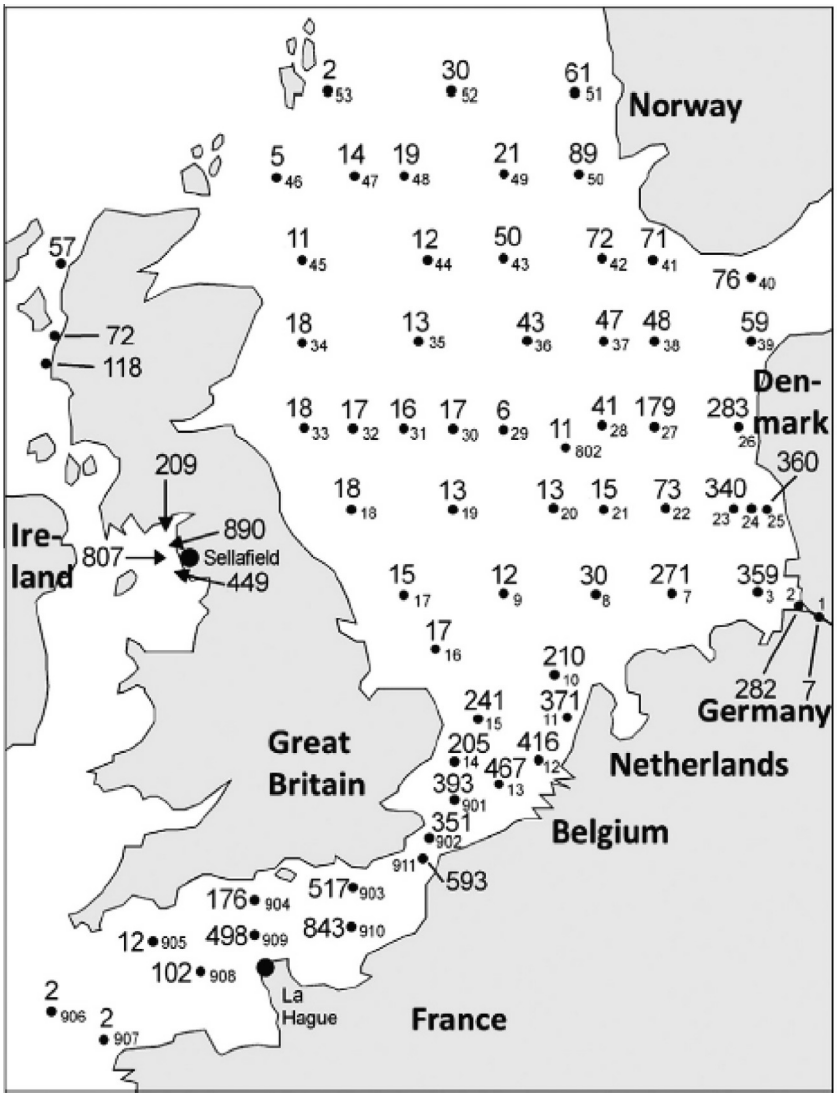

Fig. 4. Measured ^{129}I concentrations (mBq/m^3) in surface waters. North Sea samples were collected in 2005. The smaller numbers are sampling station numbers and the larger ones the corresponding measured concentrations (taken from Michel et al., 2012).

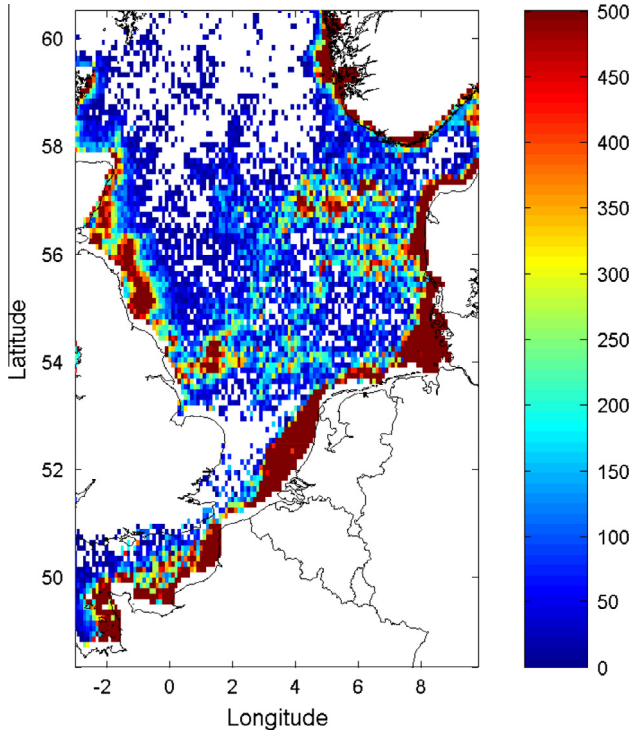

Fig. 5. Calculated ^{129}I concentrations (mBq/m^3) in surface waters (mean value over a surface layer with thickness 100 m) of the North Sea for year 2005.

In practice, it is multiplied by *RAN* to obtain the real size at a given time and for a given particle. Similarly, the maximum size of the vertical step is (Proctor et al., 1994; Hunter, 1987; Periáñez and Elliott, 2002):

$$D_v = \sqrt{2K_v\Delta t} \tag{3}$$

given either towards the sea surface or the sea bottom. K_h and K_v are the horizontal and vertical diffusion coefficients respectively and Δt is time step. A constant typical value of 1.0×10^{-3} m^2/s (Elliott et al., 2001) has been used for K_v. It has been tested that model results are little sensitive to this parameter. Actually, most of the ^{129}I remains in the surface layer, as has been found in other modelling studies (Orre et al., 2009). The Smagorinsky's scheme (Cushman-Roisin and Beckers, 2011) has been adopted to describe the horizontal diffusivity.

Radioactive decay can also be simulated by a stochastic method (Periáñez and Elliott, 2002), although this process is neglected given the very long half-life of ^{129}I (1.6×10^7 years) in comparison with simulated times (47 years). Although radionuclide adsorption/desorption reactions between water and sediments can also be simulated using a stochastic method (Periáñez and Elliott, 2002), these processes are also neglected here since iodine is conservative in seawater, thus remaining in solution.

The considered domain in the north Atlantic extends from 50°W to 25°E in longitude and from 45.1°N to 75.1°N in latitude (Fig. 1). Water circulation for the period of interest 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 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, the surface water circulation for January 2008 is presented in Fig. 2. The Norwegian Coastal Current (NCC), flowing along the Norwegian coast to the north, is clearly seen. The East Greenland Current (EGC) flows southwards along the east Greenland coast. Finally, the North Atlantic Current (NAC), deriving from the Gulf Stream, is apparent in the central Atlantic with a number of eddies and meanders.

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

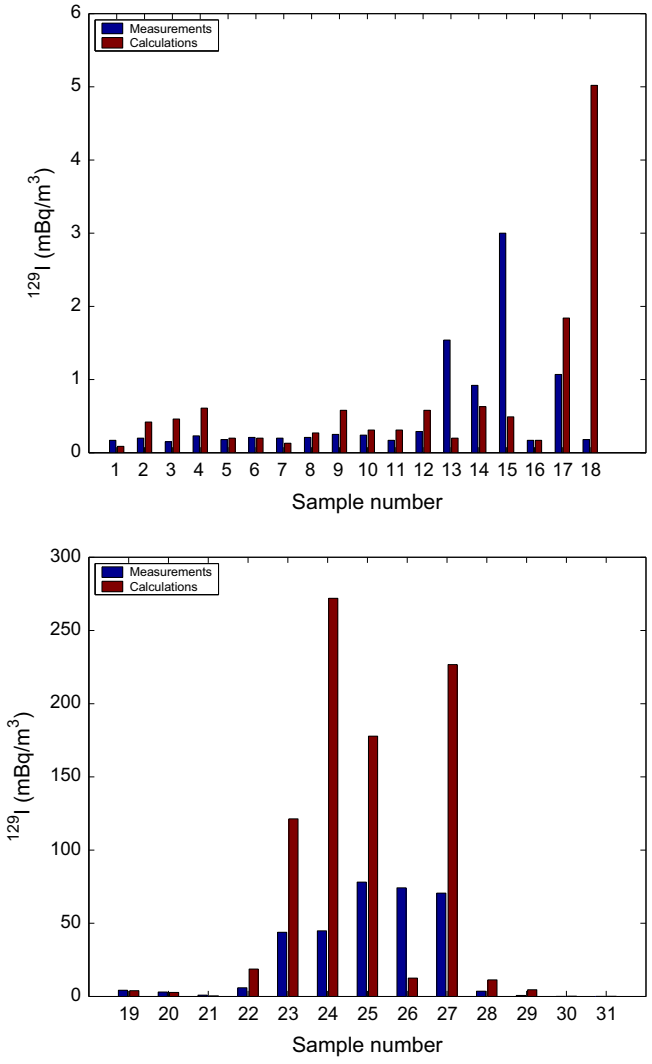

Fig. 6. Calculated (over a 100 m thick layer) and measured ^{129}I concentrations (mBq/m^3) in surface waters at points indicated in Fig. 1 for samples collected in 2010 and 2012 (1–18; top panel. From Gómez-Guzman et al. (2013) and López-Gutiérrez et al. (2013)) and 1999 (19–31; bottom panel. From Alfimov et al. (2004)).

Annual ^{129}I releases from Sellafield and La Hague, presented in Fig. 3, are introduced from 1966 to 2012, which is the simulation period. These releases define the number of Bq (or atoms) corresponding to each released particle.

3. Results and discussion

Model results have been first compared with available measurements to test its performance. Michel et al. (2012) present a map with ^{129}I concentrations in the surface waters of the North Sea and English Channel for samples collected in 2005 (Fig. 3 in their paper, which is reproduced in Fig. 4). The authors do not give the exact location of sampling points. Thus, a detailed comparison cannot be done. However, the overall calculated distribution

pattern of ^{129}I is in agreement with observations. This calculated distribution is presented in Fig. 5. Surface concentrations are calculated as the mean value over a 100 thick surface layer. The thickness of the surface mixed layer in the ocean typically ranges from 25 to 200 m (Pickard and Emery, 1982). Thus, we provide concentrations in the surface as an average over a 100 m thick layer, which may be representative of the mean value of the surface mixed layer thickness.

Concentrations about 500 mBq/m^3 are obtained in the English Channel, in agreement with Michel et al. (2012). A strong concentration gradient is apparent in Denmark, with quickly decreasing concentrations as moving offshore into the North Sea. This effect is also found in Michel et al. (2012), although calculated concentrations are slightly overestimated by the model. A similar pattern is obtained along the UK eastern coast. However, this cannot be seen in the measurements of Michel et al. (2012) because samples were not collected close to the coast, but tens of km offshore. Since concentrations decrease very fast as moving offshore, probably these high concentrations calculated by the model along the British coast cannot be appreciated in Michel et al. (2012). In the central North Sea, measured concentrations generally are of the order of 10^1 mBq/m^3. Model calculations are in agreement with this (Fig. 5). Measured ^{129}I concentrations within the Irish Sea are not totally reproduced by the model, which must be due to the relatively low resolution of JAMSTEC currents. Spatial structure of currents may not be adequately represented in semi-enclosed basins with narrow connections with the open ocean. The same situation occurs in the Danish Straits, as will be shown below. Nevertheless, the focus of this paper is the open Atlantic Ocean, not shelf seas. Moreover, the Irish Sea is very sensitive to the discharge timing. Thus, modelled and measured results can be very different here, depending of the sampling date. Also, it must be considered that mean annual discharges are being used in the model, which affects model accuracy in the near field.

Water samples have also been collected around Iceland, in the Irminger Sea and in a transect from Scotland to Iceland (Fig. 1) in 2010 (Gómez-Guzman et al., 2013) and 2012 (López-Gutiérrez et al., 2013). A comparison between measured and calculated ^{129}I surface concentrations is presented in Fig. 6. Generally speaking, calculated concentrations in the UK-Iceland transect are in good agreement with observations. The model underestimates concentrations in some sampling points in the area of Iceland (points 13, 14 and 15). Salinity and temperature measurements indicate that North Atlantic Water is present here (Gómez-Guzman et al., 2013). Thus, the origin of this ^{129}I cannot be attributed to the EGC, which eventually could be transporting radionuclides from the Arctic back to the Atlantic. Instead, some eddies or episodic currents which are not reproduced by the mean monthly water circulation could be transporting patches of Sellafield releases to this area.

An overestimation about one order of magnitude is apparent in the Irish Sea sample, which may be due to reasons mentioned above. However, model results agree with measurements in the Celtic Sea area.

Some profiles in the water column were also measured in the area of Iceland (Gómez-Guzman et al., 2013), showing decreasing concentrations with water depth. The model does not calculate significant concentrations down the water column. As commented above, it has been found in other modelling studies (Orre et al., 2009) that most of the ^{129}I released from Sellafield and La Hague remains in the surface layer. Thus, ^{129}I measured in deep waters in the area of Iceland could be coming from the Arctic. The model cannot simulate this transport from the Arctic back to the Atlantic since the former is not within the considered domain (the Arctic is not included in JAMSTEC hydrodynamic calculations).

Samples were also collected in a transect from the North Atlantic to the Baltic Sea in 1999 (Alfimov et al., 2004). A comparison of model results with measurements is also presented in Fig. 6. The model is overestimating concentrations in the Danish Straits. This may be due to the relatively coarse resolution of the model, which does not reproduce adequately the narrow straits and passages in these waters. This makes particles to remain trapped in the area. Nevertheless, the overestimation is not larger than a factor 5.

As an example, the calculated ^{129}I concentration in surface water for year 2000 is presented in Fig. 7. Radionuclides follow

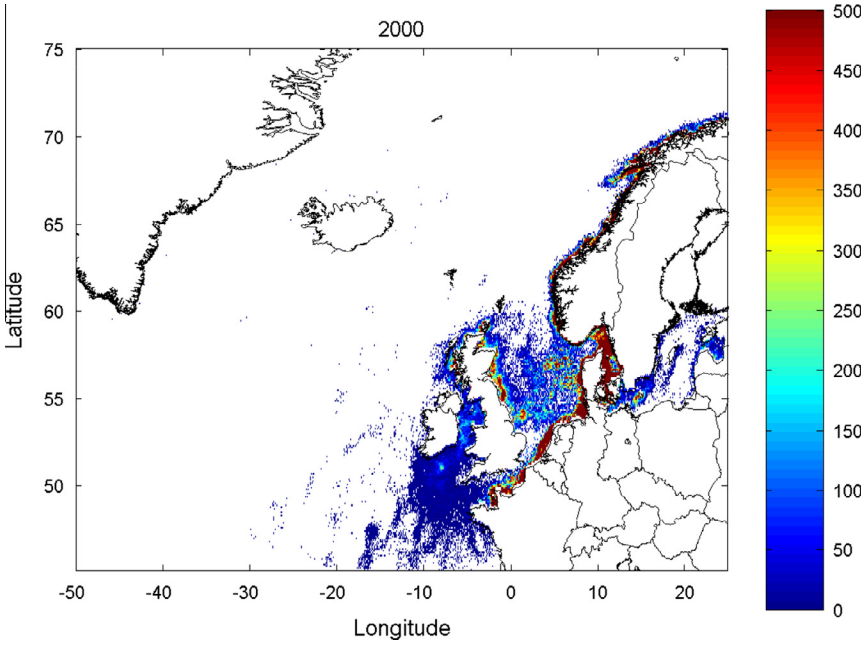

Fig. 7. Calculated ^{129}I concentrations (mBq/m^3) in surface waters (100 m layer) in year 2000. The maximum of the colour scale has been set to 500 mBq/m^3. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

the well known transport pathways (Orre et al., 2009; Povinec et al., 2003). Thus, Sellafield releases travel around Scotland and enter the North Sea. Here they join La Hague releases and then travel along Norway coast with the NCC. Part of the activity previously enters the Baltic Sea. Strong gradients are obtained across the NCC, which has already been found in the case of ^{137}Cs, and is also in agreement with previous ^{129}I simulations (Orre et al., 2009). In addition, some of the Sellafield released radionuclides are transported to the south, as has been found for ^{137}Cs (Povinec et al., 2003). Then part of this activity enters the English Channel and, from here, the North Sea.

A main contribution of this work is to provide a quantitative analysis on the fate of ^{129}I released from Sellafield and La Hague. First, the activity inventory in the Arctic Ocean has been evaluated. Particles in the Arctic are defined as those which leave the model through the north and north-east open boundaries. As commented before, transport of radionuclides back from the Arctic to the Atlantic cannot be simulated. Thus, results should be considered as an upper limit. Model results are presented in Figs. 8–10 for radionuclides respectively released from Sellafield, La Hague, and considering both sources. Note that, given the Lagrangian nature of the model, different labels can be assigned to particles released from

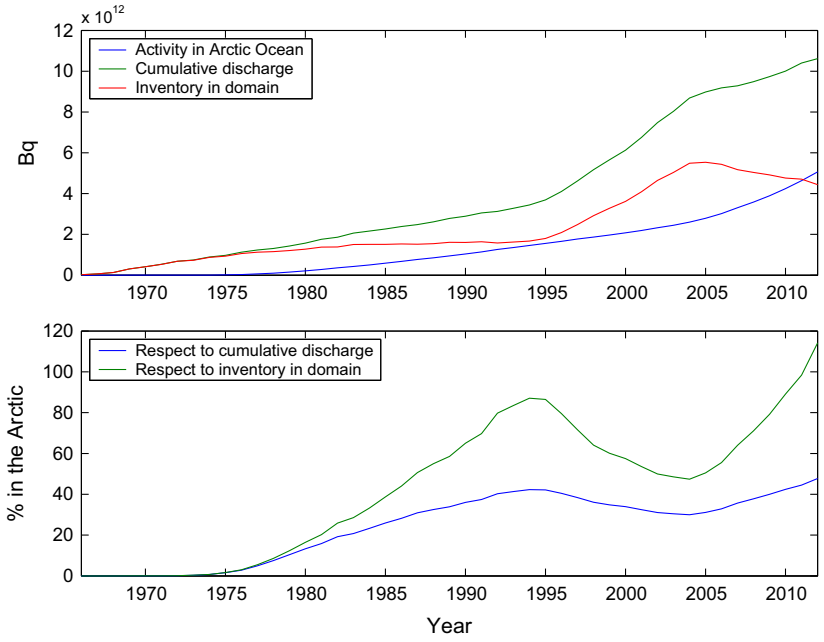

Fig. 8. Top: total inventory of ^{129}I in the Arctic, Sellafield cumulative discharge and inventory in the model domain for releases from Sellafield. Bottom: fraction of the Arctic inventory with respect to the cumulative discharge and to the inventory in the domain.

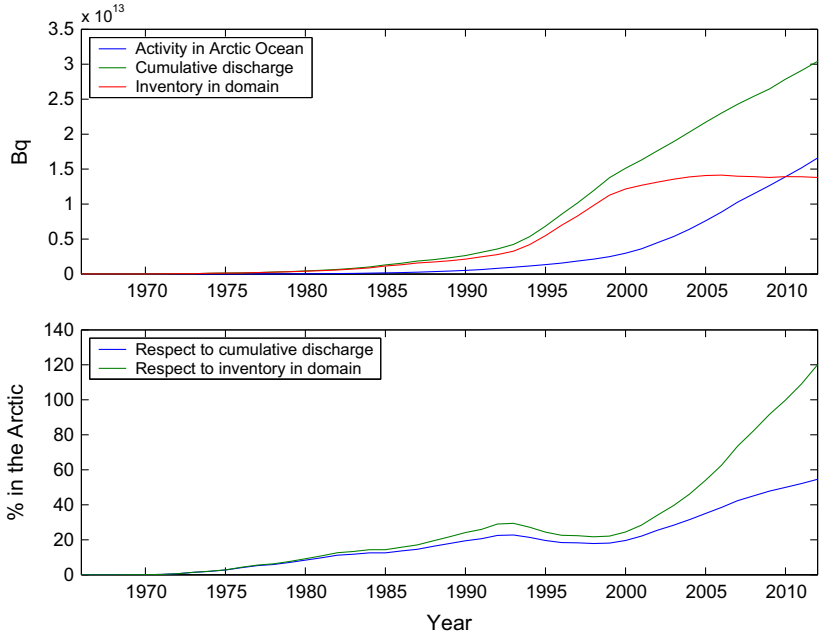

Fig. 9. Same as Fig. 8 but for releases from La Hague.

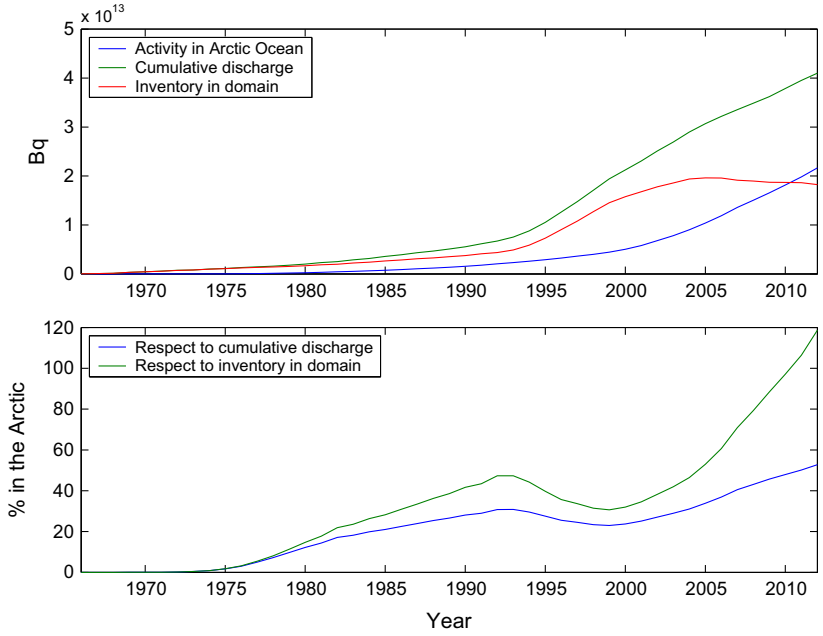

Fig. 10. Same as Fig. 8 but for releases from Sellafield and La Hague together.

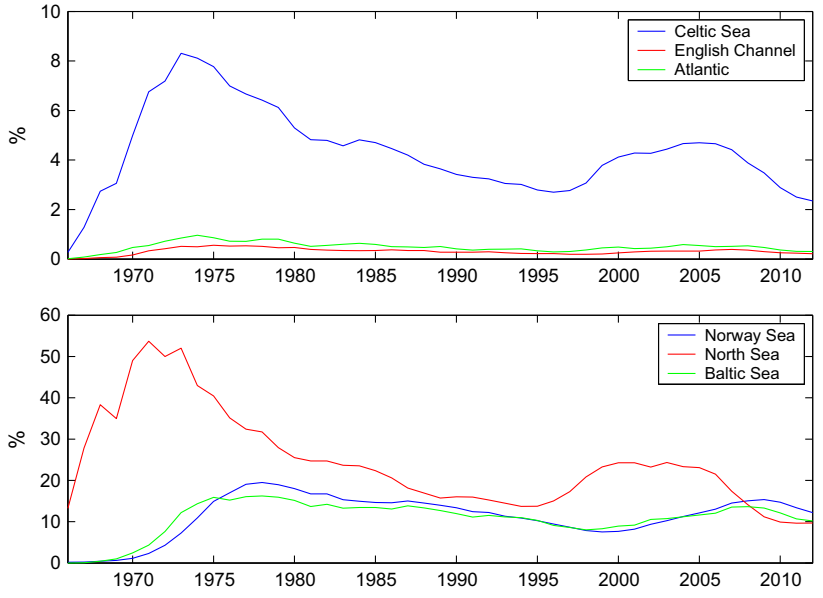

Fig. 11. Fractions of ^{129}I in several basins with respect to the cumulative discharge for releases from Sellafield.

La Hague and Sellafield. Thus, a detailed analysis may be carried out since the origin of each particle in the computational domain is known.

Radionuclide inventory in the Arctic increases monotonically along the simulation period. About 48% of the total releases from Sellafield are within the Arctic Ocean at the end of the simulation (2012). In the case of La Hague releases this fraction is slightly higher (55%) since most of the activity is transported to the east, towards the North Sea. However, part of Sellafield releases are transported south, to the Celtic Sea. There is a higher activity input to the Arctic Ocean than the activity present within the model domain (about 120%), as may be seen in Figs. 8–10. It can be concluded that 5.1 and 16.6 TBq of ^{129}I have been introduced in the Arctic from Sellafield and La Hague respectively until 2012 (Figs. 8 and 9).

The total supply of ^{129}I from both plants to the Arctic in 1993 was estimated to be $\geqslant$ 7 TBq (Kershaw and Baxter, 1995). But this figure essentially represents 100% of the cumulative discharge to that time, and seems to be largely overestimated. The calculated inventory in the Arctic for that time with the present model results 2.1 TBq (Fig. 10), which is 31% of the cumulative discharge from both plants until then. In the case of ^{90}Sr, which is also conservative in seawater, it was estimated that 30% of the Sellafield release was transported into the Barents Sea (Kershaw and Baxter, 1995).

Inventories in the North Atlantic, including shelf seas (i.e., the model domain), are 4.4 and 13.8 TBq, coming from Sellafield and La Hague respectively, in 2012 (Figs. 8 and 9).

The fractions of radionuclides in different regions of the North Atlantic (with respect to the cumulative discharges) are presented

in Figs. 11 and 12 for Sellafield and La Hague releases respectively. These regions are indicated in the map of Fig. 1. At recent times, between 10% and 15% of the releases are present in the North Sea, Baltic Sea and Norway Sea, for both Sellafield and La Hague releases. In the case of La Hague, the inventory in the English Channel is about 3% of the discharge, and in the Celtic Sea and open Atlantic it is negligible (Fig. 12). In the case of Sellafield, about 2% of the discharge is in the Celtic Sea in 2012 and less than 1% is present in both the English Channel and open Atlantic. This is similar to previous results for ^{137}Cs: it has been found (Povinec et al., 2003) that, on average, 1% of Sellafield releases go through the English Channel.

The model has been applied to calculate the mean age of the radioactivity content in the model domain. The concept of age is widely used in ocean sciences to highlight the inherent time scales of a system associated with the advection and diffusion of matter. It has been used for two main purposes: to estimate the ventilation rate of ocean basins and to infer horizontal circulation from tracer dispersion. In the last case the age is defined as the time elapsed since a tracer was released in the sea from a point source. As described in detail by Deleersnijder et al. (2001), the age is a Lagrangian concept: one just needs to attach a "clock" to each particle. In our case, the clock is initialized when the particle is released from Sellafield or La Hague plants. At the end of the simulation, the ages indicated by the clocks attached to the particles in the ocean are read. The "age-averaging hypothesis" (Deleersnijder et al., 2001) is applied to evaluate the mean age, which reads that the mean age of a set of particles (in our case, particles which are within each grid cell) is defined as the mass-weighted arithmetic average of the ages of the particles considered. The mass of each

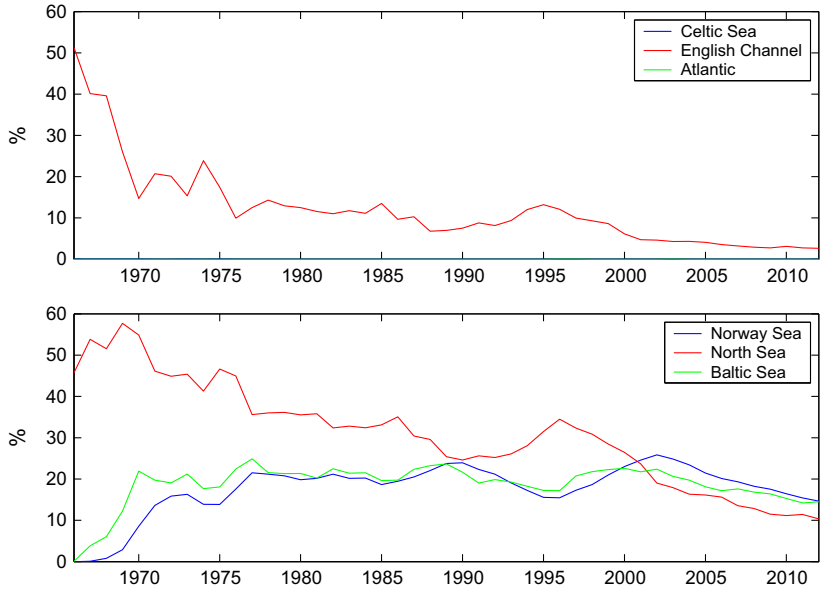

Fig. 12. Fractions of ^{129}I in several basins with respect to the cumulative discharge for releases from La Hague.

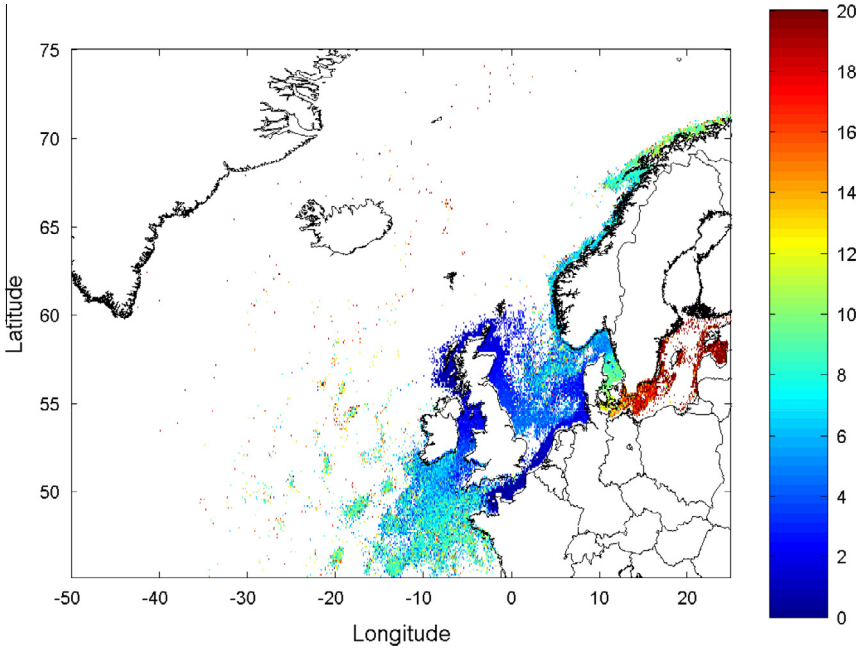

Fig. 13. Calculated ^{129}I mean ages (years) over the model domain.

particle corresponds to its radioactivity content in the present application. It must be pointed out that age is a different concept from transit time, which are well defined for this area [for instance in Povinec et al., 2003)] and are not calculated again in this work.

Calculated mean ages over the model domain are presented in Fig. 13. Obviously, tracer age increases with increasing distance from the source (Sellafield and La Hague). Mean age of tracers in the North Sea is about 5 years, while it is larger in the Celtic Sea (approaching to some 10 years in the south). Mean age in the Baltic is significantly longer (more than 20 years). This is due to the fact that particles, once they enter the Baltic, remain there for a significant time due to the limited water exchange with the North Sea. Actually, residence times estimated for the Baltic range from 11 to 30 years (Lepparanta and Myrberg, 2009). Mean age increases along the NCC, reaching more than 10 years in the area of North Cape. In the open Atlantic Ocean, tracer ages are of the order of 20 years.

Given the Lagrangian nature in the model, the origin of each particle is known. Thus, mean ages for releases coming from Sellafield and La Hague can be calculated separately. These are shown in Fig. 14. These pictures are also useful to see in detail the different fates of Sellafield and La Hague releases, already outlined in Fig. 7. The lasts follow a very straight path towards the NCC, although some is lost into the Baltic Sea. Some of the Sellafield releases enter the North Sea from the English Channel and some from the north (around Scotland). These two fractions contribute differently to the mean ages of Sellafield releases in the North Sea. Particles coming from the English Channel have mean ages around 8 years, while particles coming from Scotland have mean ages about 2 years. As a result of the mixing of these two fractions of particles, mean ages in the southern NCC (at 60°N latitude) is about 9 years. On the other hand, mean age in this area for particles coming from La Hague is about 5.5 years. Consequently, mean ages for Sellafield releases are about 3.5 years

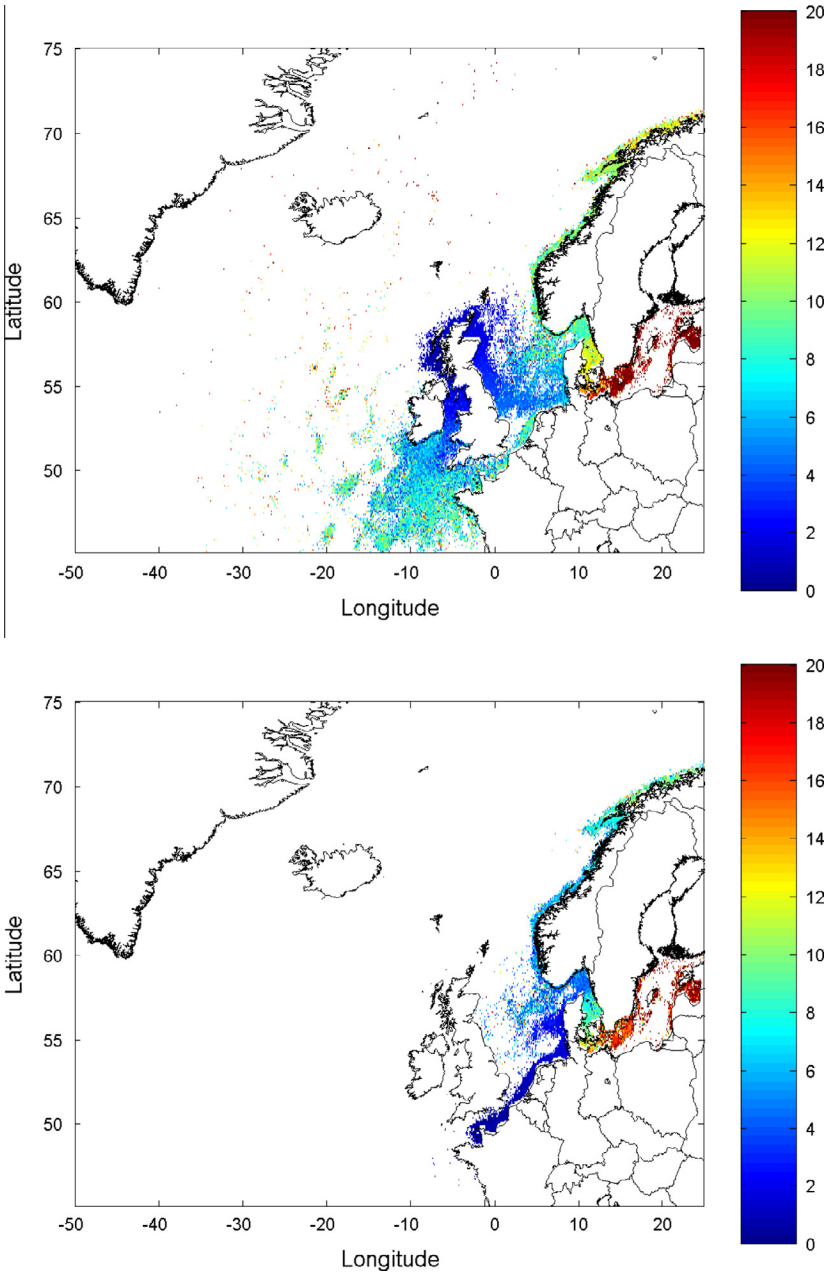

Fig. 14. Calculated ^{129}I mean ages (years) over the model domain. Top: Sellafield releases. Bottom: La Hague releases.

larger than for La Hague releases in this zone. Since releases from Sellafield and La Hague travel together to the Arctic following the NCC, this age difference will be maintained during such trip. When releases from both plants are considered, mean age in the southern NCC is (Fig. 13) about 7 years. This number is not the simple average value between Sellafield and La Hague particle ages, since mass-weighted arithmetic means are used to calculate mean ages (Deleersnijder et al., 2001).

4. Conclusions

A Lagrangian model to simulate the dispersion of ^{129}I in the northern Atlantic Ocean has been developed. It uses pre-computed current fields (monthly means values) and has been validated through the comparison of measured ^{129}I concentrations with calculated values. New estimations of ^{129}I inventories in the Arctic Ocean and European seas have been provided.

It has been found that about 48% and 55% of the total releases from Sellafield and La Hague, respectively, have entered the Arctic Ocean at the end of the simulation (2012). Some 5.1 and 16.6 TBq of ^{129}I have been introduced in the Arctic from Sellafield and La Hague respectively until 2012. These figures are lower than previous estimations based on field measurements of ^{129}I concentrations. Real inventories in the Arctic must be even lower than our estimations, since the transport of radionuclides from the Arctic back to the Atlantic Ocean is not simulated.

The presence of ^{129}I in European shelf seas has been determined as well. At recent times, between 10% and 15% of the releases are present in the North Sea, Baltic Sea and Norway Sea, for both Sellafield and La Hague releases. In the case of Sellafield there is also a significant southward transport, to the Celtic Sea. It has been found that there is a higher content of ^{129}I in shelf seas than in open Atlantic Ocean waters.

Tracer ages have been finally calculated. They increase with increasing distance from the source (Sellafield and La Hague). Mean age of tracers in the North Sea is about 5 years, while in the Baltic is significantly longer (more than 20 years). Mean age increases along the NCC, reaching more than 10 years in the area of North Cape. In the open Atlantic Ocean, tracer ages are of the order of 20 years. Mean ages for Sellafield releases are about 3.5 years larger than for La Hague releases in the North Sea and subsequent areas through which radionuclides travel.

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