Science of the Total Environment 621 (2018) 376–386

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Recent evolution of ^{129}I levels in the Nordic Seas and the North Atlantic Ocean

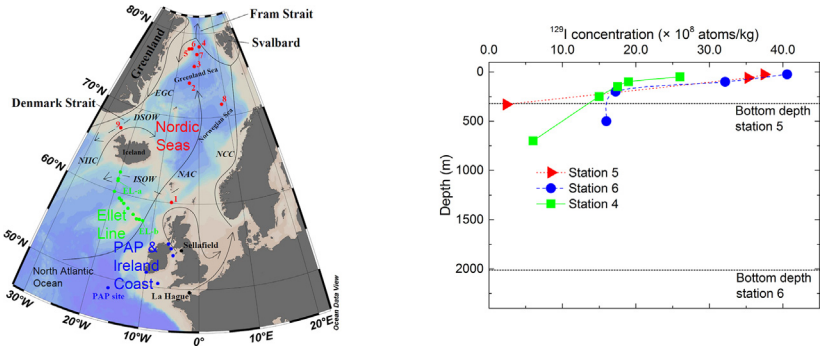
Carlos Vivo-Vilches [a,b,*], José María López-Gutiérrez [a,b], Raúl Periáñez [c], Charlotte Marcinko [d], Frédéric Le Moigne [e], Paul McGinnity [f,1], Juan Ignacio Peruchena [b], María Villa-Alfageme [g]

[a] Departamento de Física Aplicada I, Escuela Politécnica Superior, Universidad de Sevilla, Virgen del África 7, 41011 Seville, Spain
[b] Centro Nacional de Aceleradores (CNA) (Universidad de Sevilla, CSIC, Junta de Andalucía), Thomas Alva Edison 7, 41092 Seville, Spain
[c] Departamento de Física Aplicada I, Escuela Técnica Superior de Ingeniería Agronómica, Universidad de Sevilla, Ctra. Utrera km 1, 41013 Seville, Spain
[d] National Oceanography Centre (NOC), European Way, Southampton SO14 3ZH, United Kingdom
[e] GEOMAR, Wischhofstrasse 1-3, D-24148 Kiel, Germany
[f] Environmental Protection Agency, 3 Clonskeagh Square, Dublin 14, Ireland
[g] Departamento de Física Aplicada II, Escuela Técnica Superior de Ingeniería de Edificación, Universidad de Sevilla, Reina Mercedes 4A, 41012 Seville, Spain

HIGHLIGHTS

- Update of ^{129}I distribution in Central North Atlantic (NA) and the Nordic Seas.
- Evolution of ^{129}I from 2002 to 2012 is associated to changes in releases from NFRPs.
- Results suggest deep water formation in the eastern area of the Nordic Seas.
- ^{129}I from NFRPs partly reaches Central NA and are not carried northwards by the NCC.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 28 June 2017
Received in revised form 23 November 2017
Accepted 23 November 2017
Available online xxxx

Editor: D. Barcelo

Keywords:
AMS
^{129}I
Nordic Seas
North Atlantic Ocean
Deep water formation

ABSTRACT

Most of the anthropogenic radionuclide ^{129}I released to the marine environment from the nuclear fuel reprocessing plants (NFRP) at Sellafield (England) and La Hague (France) is transported to the Arctic Ocean via the North Atlantic Current and the Norwegian Coastal Current. ^{129}I concentrations in seawater provides a powerful and well-established radiotracer technique to provide information about the mechanisms which govern water mass transport in the Nordic Seas and the Arctic Ocean and is gaining importance when coupled with other tracers (e.g. CFC, ^{236}U).

In this work, ^{129}I concentrations in surface and depth profiles from the Nordic Seas and the North Atlantic (NA) Ocean collected from four different cruises between 2011 and 2012 are presented. This work allowed us to i) update information on ^{129}I concentrations in these areas, required for the accurate use of ^{129}I as a tracer of water masses; and ii) investigate the formation of deep water currents in the eastern part of the Nordic Seas, by the analysis of ^{129}I concentrations and temperature-salinity (T-S) diagrams from locations within the Greenland Sea Gyre. In the Nordic Seas, ^{129}I concentrations in seawater are of the order of 10^9 at·kg^{-1}, one or two orders of magnitude higher than those measured at the NA Ocean, not so importantly affected by the releases from the

* Corresponding author at: Centro Nacional de Aceleradores (CNA) (Universidad de Sevilla, CSIC, Junta de Andalucía), Thomas Alva Edison 7, 41092 Seville, Spain.
E-mail address: cvivo@us.es (C. Vivo-Vilches).
[1] Currently at: IAEA Environment Laboratories, 4a, Quai Antoine 1er, 98000, Monaco.

https://doi.org/10.1016/j.scitotenv.2017.11.268

NFRP. ^{129}I concentrations of the order of 10^8 atoms·kg^{-1} at the Ellet Line and the PAP suggest a direct contribution from the NFRP in the NA Ocean.
An increase in the concentrations in the Nordic Seas between 2002 and 2012 has been detected, which agrees with the temporal evolution of the ^{129}I liquid discharges from the NFRPs in years prior to this. Finally, ^{129}I profile concentrations, ^{129}I inventories and T-S diagrams suggest that deep water formation occurred in the easternmost area of the Nordic Seas during 2012.

1. Introduction

^{129}I is a long-lived radionuclide ($T_{1/2} = 15.7 \times 10^6$ years) with a strongly increasing presence in the environment since the beginning of the nuclear era. Natural processes – the cosmogenic production by spallation reactions with Xe in the atmosphere and the fission of U isotopes – result in $^{129}I/^{127}I$ atom ratios of 10^{-13} and 10^{-12} respectively (Fabryka-Martin et al., 1987). However, the anthropogenic activities (including fallout from nuclear weapons tests and discharges from nuclear power plants and, especially, nuclear fuel reprocessing plants) have resulted in $^{129}I/^{127}I$ atom ratios several orders of magnitude higher. The use of ^{129}I as an oceanographic tracer has been extensively described in previous works (Fan et al., 2013; He et al., 2013). The effectiveness of this radionuclide for such purposes is based on three important factors. Firstly, due to its long half-life, it remains in the environment for many years. Secondly, it is strongly conservative, i.e., it remains dissolved in seawater for a long time. Finally, the source terms for the main discharges of ^{129}I into the environment for the last 50 years are well documented (AREVA, 2013; López-Gutiérrez et al., 2004; Sellafield Ltd., 2014). The main releases of ^{129}I to the marine environment are liquid discharges from the two main nuclear fuel reprocessing plants (NFRP) in Europe: Sellafield (United Kingdom) and La Hague (France), which as of 2012 have released totals of 1634 kg and 4677 kg of ^{129}I respectively (Daraoui et al., 2016).

Recently, the effectiveness of ^{129}I as a radiotracer has been further enhanced because of the development of its use in combination with another long-lived radionuclide, ^{236}U, which is also released from the NFRPs. The use of $^{129}I/^{236}U$ ratio as a complementary tracer is a powerful approach for analyzing Arctic Ocean water mass behaviour in particular (Casacuberta et al., 2016; Christl et al., 2015).

The great majority of the ^{129}I discharged into the ocean has been directly released to the Irish Sea and the English Channel. Previous studies (Alfimov et al., 2006; Alfimov et al., 2004c) showed that the majority of these releases is transported through the North Sea to the Arctic Ocean along the Norwegian coast by the Norwegian Coastal Current (Villa et al., 2015). After a transit time of approximately five years in the Arctic Ocean (Orre et al., 2010; Smith et al., 2011) most of the ^{129}I is transported Southwards through the Fram Strait to the Greenland Sea (Alfimov et al., 2004c; Gómez-Guzmán et al., 2013). Furthermore, part of the ^{129}I gets involved into longer loops in the Arctic Ocean following alternative transport pathways (Karcher et al., 2012; Smith et al., 1998).

^{129}I is a particularly good tracer for circulation studies in the Arctic. e.g. Karcher et al. (2012) used model and experimental ^{129}I data from the Arctic Ocean (mid-1990s and 2000s) to illustrate the changes in oceanic circulation which occurred during that time, suggesting the development of a new Atlantic Water circulation scheme involving a separation between flow at intermediate depths in the Eurasian and Canada Basins. Orre et al. (2010) simulated ^{129}I in the North Atlantic using a OGC Model to bring insight into time-scales of tracer transport in this region were obtained. Finally, Smith et al. (2005) and Villa et al. (2015) obtained mean ages and transit times in the Nordic Seas region making use of ^{129}I concentrations.

The Nordic Seas region may crucially influence the global climate as a result of the exchange of heat that occurs between the cold Arctic Ocean and the warm North Atlantic Ocean during the formation of the deep waters in specific areas of the Nordic Seas (Open University, 2001). New data are of fundamental importance for tracing transport pathways and features of the water masses that drive the formation of North Atlantic Deep Waters (NADW). The specific location of this formation remains a topic of discussion (Hansen and Østerhus, 2000). The most recent studies support the theory that part of the NADW are formed in the eastern Nordic Seas, that is, the easternmost part of the Greenland Sea and the Norwegian Sea (Latarius and Quadfasel, 2016).

^{129}I is an ideal tracer for this purpose (Alfimov et al., 2013). The most up-to-date studies providing information on the levels of this radionuclide in the Nordic Seas and Central North Atlantic have been determined from samples collected between 2001 and 2005. Casacuberta et al. (2016) recently published updated ^{129}I concentrations for the Arctic Ocean (Canada, Makarov and Eurasian Basins). Some other studies, such as Gómez-Guzmán et al. (2013), provide updated data on levels of ^{129}I in the Irminger Basin. However, there is no up to date, comprehensive data compilation of ^{129}I concentrations either in the North Atlantic Ocean or in the Nordic Seas. This information is key for the accurate use of this radionuclide as a tracer in these areas.

In this work the distribution of ^{129}I in the Nordic Seas is analyzed, the majority of the samples analyzed are from stations located in the Norwegian Sea and the Greenland Sea. These seas connect the North Atlantic and the Arctic Oceans, thus, as previously described, they are important areas in which to monitor possible changes in the thermohaline circulation, since it is related to the North Atlantic Deep Water (NADW) formation.

The results for ^{129}I concentrations in surface seawater samples for four areas in the Nordic Seas and the North Atlantic Ocean are presented. Additionally, 11 specific depth profiles have been analyzed. The main objective of this study was to determine the current levels of ^{129}I due to NFRP discharge in these areas and to apply this data to the study of the oceanic currents. The evolution of ^{129}I concentrations in currents flowing in and out of the Nordic Seas have been analyzed and the formation of overflow waters in the Nordic Seas have been investigated through measured vertical distributions and inventories of this radionuclide.

2. Methods

2.1. Sampling

^{129}I concentrations and $^{129}I/^{127}I$ atom ratios were measured in samples from four cruises which were undertaken in 2010, 2011 and 2012 (Fig. 1).

Samples from the PAP site were sampled on board the RRS James Cook during cruise JC071 from Glasgow to the PAP site, from 29th April to 12th May 2012. The Porcupine Abyssal Plain Sustained Observatory (PAP-SO) is a multidisciplinary observatory in the North Atlantic. These samples were collected in one of the regular cruises that take place to the site to collect equipment time series datasets. Samples at the Ellet line were sampled on board RRS Discovery (cruise D379 in August 2012). The Ellet Line is a route of long term oceanographic observations that links Scotland and Iceland through the Rockall Plateau and the Iceland Basin, separating the Nordic Seas from the North Atlantic. During the Ellet cruises, regular samplings are made to assess climate stability.

Greenland, Norwegian and Barents Seas were sampled on board James Clark Ross (British Antarctic Survey) during the UK's Ocean Acidification program, from 12th June to 1st July 2012.