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New insights on the role of sea ice in intercepting atmospheric pollutants using ^{129}I

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ABSTRACT

Measurements of ^{129}I carried out on sea ice samples collected in the central Arctic Ocean in 2007 revealed relatively high levels in the range of 100–1400 $\times$ 10^7 at L^{-1} that are comparable to levels measured in the surface mixed layer of the ocean at the same time. The $^{129}I/^{127}I$ ratio in sea ice is much greater than that in the underlying water, indicating that the ^{129}I inventory in sea ice cannot be supported by direct uptake from seawater or by iodine volatilization from proximal (nearby) oceanic regimes. Instead, it is proposed that most of the ^{129}I inventory in the sea ice is derived from direct atmospheric transport from European nuclear fuel reprocessing plants at Sellafield and Cap La Hague. This hypothesis is supported by back trajectory simulations indicating that volume elements of air originating in the Sellafield/La Hague regions would have been present at arctic sampling stations coincident with sampling collection.

1. Introduction

Oceanic discharges of anthropogenic radionuclides from European nuclear fuel reprocessing plants (mainly Sellafield and La Hague) have introduced a suite of radionuclide contaminants to the marine environment (eg. ^{137}Cs, ^{90}Sr, ^{99}Tc, ^{236}U, Pu isotopes and ^{129}I (AMAP, 2004)) that are useful as tracers for detailed investigations of ocean processes. These studies include surface circulation and mixing in the Nordic Seas (Alfimov et al., 2013), surface and intermediate circulation in the Arctic Ocean (Karcher et al., 2012; Smith et al., 2011) and in the North Atlantic (Casacuberta et al., 2014), and the descent of the dense overflows from the Greenland and Norwegian Seas into the deep North Atlantic (Orre et al., 2010).

The Arctic Ocean is covered by seasonal sea ice that plays an important role in the global and regional climate system, as well as in the oceanic circulation. Sea ice is formed during winter, mostly on the shallow continental shelves of the Laptev, the Kara and the Barents Seas (Aagaard, 1981). The drifting of sea ice is governed by the Transpolar Drift (TPD) over the Eurasian Basin and the anticyclonic Beaufort Gyre in the Canadian Basin (Barrie et al., 1998). Different chemical species, including radionuclides, can be incorporated into the sea ice through direct input (uptake from seawater during its formation and interception of atmospheric dry and wet deposition). For example, Cámara-Mor et al. (2011) determined that Arctic sea ice can intercept a significant fraction 30% ± 18% of the atmospheric inputs of the cosmogenic radionuclide ^{7}Be, depending on the sea ice coverage, and suggested that other contaminants could be similarly accumulated via atmospheric pathways. Chemical substances can also accumulate in sea ice through indirect inputs by uptake from sediments and biota such as algae entrained into the ice. As sea ice reaches ablation areas such as the Fram Strait and, to a lesser degree, the Canadian archipelago, it melts and releases the incorporated chemical species, including radionuclides, to the surface water (Pfirman et al., 1997; Rigor et al., 2002; Masqué et al., 2003). Hence, sea ice has been identified as playing a potentially important role in the

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redistribution and transport of those chemical species in the Arctic Ocean (Masqué et al., 2003, 2007; Cámara-Mor et al., 2010).

^{129}I is a highly soluble and long-lived ($T_{1/2} = 15.7 \times 10^6$ years) radionuclide that has been extensively discharged to the environment since the beginning of the nuclear era. Although large amounts of this radionuclide were released during atmospheric nuclear weapons tests and the Chernobyl accident (Raisbeck et al., 1995; Wagner et al., 1996; Gallagher et al., 2005), nuclear fuel reprocessing plants, especially Sellafield (UK) and La Hague (France), are the main sources responsible for recent ^{129}I increases in the environment. These plants were discharging to the sea about 20 kg year^{-1} (9.34×10^{25} atoms, 13.2 GBq) between 1965 and the beginning of the 1990's. Later, the releases increased up to 300 kg year^{-1} (1.40×10^{27} atoms, 198 GBq), mainly from La Hague (López-Gutiérrez et al., 2004; UNSCEAR, 2000). There is some evidence suggesting that contaminated Russian rivers from Siberia can also be a source of ^{129}I, but this contribution is nevertheless of minor importance for the large scale distribution of ^{129}I in the north Atlantic and the Arctic Oceans (Cooper et al., 2001; Cochran et al., 2000).

It has been shown that coastal currents transport ^{129}I from the Irish Sea and the English Channel to the North Sea and the Arctic Ocean along the Norwegian coast (Raisbeck et al., 1995) (Fig. 1). In the Arctic Ocean, ^{129}I concentrations can be more than an order of magnitude higher in Atlantic-origin waters compared to Pacific-origin water, and as a result there is a pronounced tracer front between waters of Pacific and Atlantic origin in the Arctic Ocean (Smith et al., 1998, 1999). Transit times of contaminants in surface water to the Arctic Ocean from the North Sea are 2–4 year to the Barents Sea, 9–10 year to the North Pole and greater than 14 year to the Canada Basin (Dahlgaard, 1995; Smith et al., 1999, 2011; Buraglio et al., 1999; Alfimov et al. 2004).

Due to its high solubility in seawater, ^{129}I concentrations in seawater reflect the general circulation patterns of the different water masses (Orre et al., 2010; Alfimov et al., 2004). Hence, in the Nansen basin ^{129}I is found predominantly in the upper part (0–500 m) of Atlantic-origin water, reflecting the water mass supply corresponding to the Fram Strait water branch, whereas in the Amundsen basin ^{129}I is distributed deeper, down to 1000 m, reflecting an additional input of ^{129}I labelled water from the Barents Sea (Barents Sea Branch Water) (Alfimov et al., 2004; Smith et al., 2011).

Kieser et al. (2005) reported elevated concentrations of ^{129}I in the range of $(80–140) \times 10^5$ atoms m^{-3} in air filters collected along Canada in 1981–1983. In that case, back trajectory analysis of air parcels showed transport from Europe and Russia before arriving to Canada and the Arctic Ocean, and indicated a consistent origin over nuclear processing sites east of the Urals, presumably

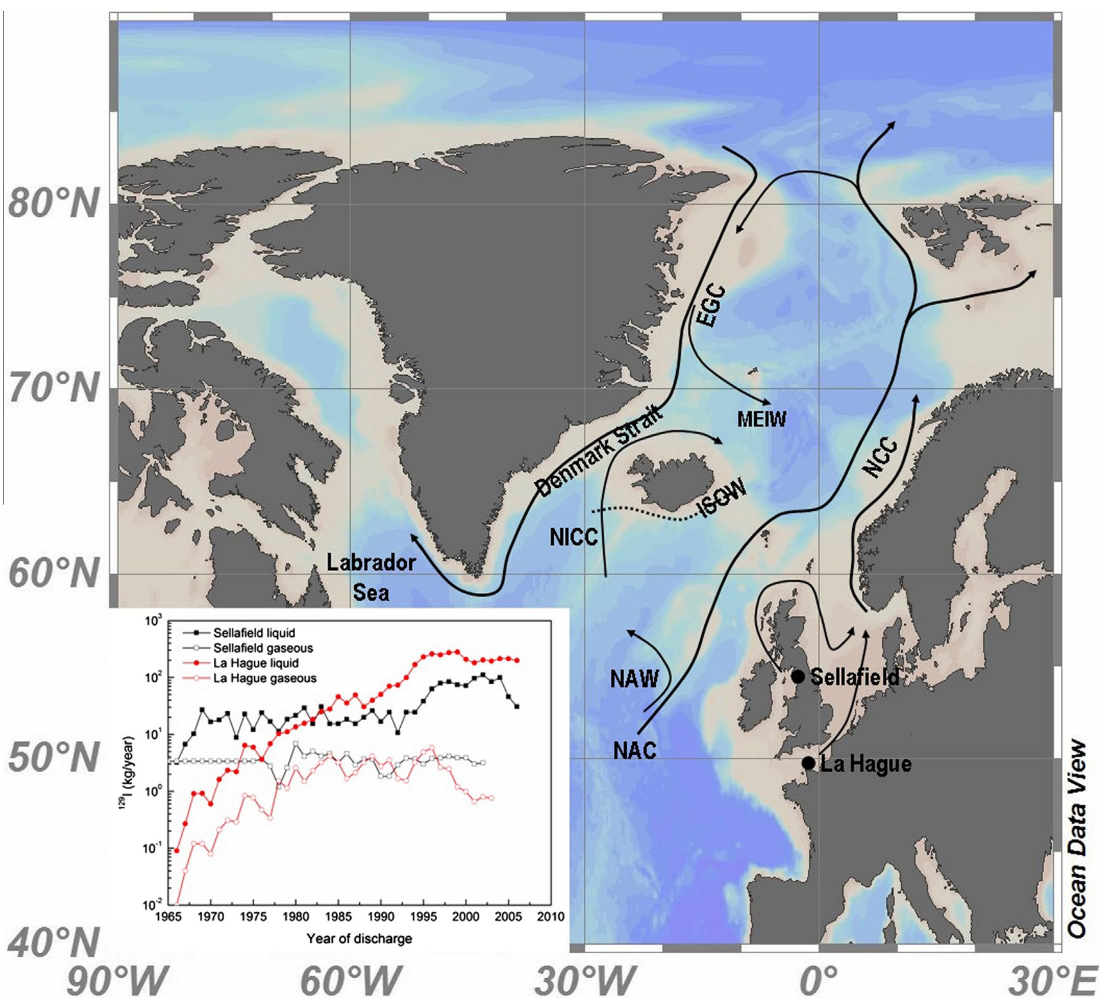

Fig. 1. Map showing the major pathways of marine discharges from Sellafield and La Hague nuclear reprocessing facilities to the Arctic and North Atlantic Oceans. Solid lines = surface currents, dashed lines = deep currents, LS = Labrador Sea, NCC = Norwegian Coastal Current, EGC = East Greenland Current, ISOW = Iceland-Scotland Overflow Water, NAC = North Atlantic Current, NAW = North Atlantic Water, NIIC = North Icelandic Irminger Current, MEIW = Modified East Icelandic Water. Inset shows history of ^{129}I liquid and gaseous discharges (kg/y) from Sellafield (black line) and La Hague (red line) Data extracted from López-Gutiérrez et al. (2004) and He et al. (2013). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)