Available online at www.sciencedirect.com
ScienceDirect
Quaternary Geochronology 3 (2008) 268–275
QUATERNARY GEOCHRONOLOGY
www.elsevier.com/locate/quageo

Measurement of iodine-129 in seawater samples collected from the Japan Sea area using accelerator mass spectrometry: Contribution of nuclear fuel reprocessing plants

Takashi Suzuki[a,*], Shoji Kabuto[a], Hikaru Amano[a], Orihiko Togawa[b]

[a]*AMS Management Section, Japan Atomic Energy Agency, Mutsu, 4-24 Minato-machi, Aomori 035-0064, Japan*
[b]*Research Group for Environmental Science, Japan Atomic Energy Agency, Tokai, Naka, Ibaraki 319-1195, Japan*

Received 5 November 2007; received in revised form 12 December 2007; accepted 20 December 2007
Available online 4 January 2008

Abstract

The concentrations of ^{129}I in seawater samples from two sites (off Sekine and the Toyama Bay) in the Japan Sea were determined by accelerator mass spectrometry. The observed concentrations exceed amounts expected from natural origin and globally distributed fallout due to nuclear weapons testing. Because the fraction of natural origin and global fallout is 2% and 8.9–13.8%, respectively, the residual more than 80% of the concentration must come primarily from nuclear fuel reprocessing plants. This result indicates a rapid distribution of ^{129}I through atmospheric transport on a global scale. The depth distribution of ^{129}I at the Toyama Bay in the Japan Sea shows that the ^{129}I maximum is in the mixed layer and that concentrations decrease with depth. The inventory of ^{129}I in water column is four times higher than that measured in the Gulf of Mexico which has almost the same depth at the Toyama Bay. This higher inventory probably reflects: (1) the rapid water sinking in the Japan Sea, (2) the difference of distance in sampling locations with respect to major ^{129}I releasing plants and (3) the strong increase in emissions from nuclear fuel reprocessing plants after the profile of the Gulf of Mexico was taken.

Keywords: Iodine; Iodine-129; The Japan Sea; Migration behavior; Nuclear fuel reprocessing plant; Anthropogenic radionuclide

1. Introduction

Iodine-129 is a long-lived radioisotope with a half-life of 1.57×10^7 years and is produced in nature by the interaction of cosmic-ray with xenon in the atmosphere and by spontaneous fission of uranium in the geosphere. About 170 kg of ^{129}I in the hydrosphere (100 kg in the ocean (Raisbeck and Yiou, 1999)) plus 56 kg of ^{129}I in the lithosphere gives a total of about 226 kg of naturally occurring ^{129}I (NCRP, 1983). During the last 60 years, the concentrations of ^{129}I in environmental samples have dramatically increased due to nuclear activities. Four major sources of releases into the environment from nuclear activities have been identified. The first one is releases during nuclear weapons testing, particularly atmospheric testing. The testing occurred the main period of 1950s and ended with the Nuclear Weapons Test Ban Treaty of 1963 (Carter and Moghissi, 1977). Routine operations of nuclear power plants are potential sources of releases, although their contributions would be minor. Nuclear accidents also released ^{129}I into the environment. The severest accident was an explosion of the Chernobyl reactor in 1986. Finally, reprocessing plants of nuclear fuels are a major source by which ^{129}I can enter the environment. From 1944 through 1972, the plutonium-production operation at Hanford Site in Washington released about 266 kg of ^{129}I into the atmosphere. The integrated discharges of ^{129}I La Hague (1975–1997) in France and Sellafield (1961–1997) in the UK were estimated to be 1640 and 720 kg, respectively, giving the overall total of 2360 kg (Raisbeck et al., 1995; Raisbeck and Yiou, 1999).

Although a commercial-based nuclear fuel reprocessing plant will be operated at the northernmost part of the main

*Corresponding author. Tel.: +81 175 45 1732; fax: +81 175 22 4213.
E-mail address: suzuki.takashi58@jaea.go.jp (T. Suzuki).

1871-1014/$ - see front matter © 2008 Elsevier Ltd. All rights reserved.
doi:10.1016/j.quageo.2007.12.004

island in Japan in the near future, the data of ^{129}I concentrations in environmental samples around Japan are very limited. Some data using Neutron Activation Analysis (NAA) were reported around the Tokai pilot plant (Muramatsu and Ohmomo, 1986; Muramatsu and Yoshida, 1995). Since the detection limit of NAA is higher than that of the accelerator mass spectrometry (AMS), we have set up an iodine beam line for the measurement of ^{129}I (Suzuki et al., 2004) which has a good performance (Suzuki et al., 2006). The ^{129}I data of soil, seaweed and milk samples around Japan were reported (Suzuki et al., 2007) and several soil data in Japan were also reported by the development of $^{129}I^{7+}$ AMS system at the University of Tokyo (Matsuzaki et al., 2007). However, there are still limited data to understand the behavior of this nuclide released in the environment. In this paper, measurement techniques for ^{129}I in seawater were developed and then discussed the origin of ^{129}I in surface seawater and finally discussed the migration behavior of iodine on the earth from the obtained depth profile.

2. Site descriptions

The Japan Sea is isolated from the Pacific Ocean and the Okhotsk Sea by four straits which have depths of less than 130 m. It is characterized by its basin-like and semi-enclosed geological structure, with a surface area of $1.01 \times 10^6\,km^2$, a mean depth of 1350 m and a maximum depth of 3796 m (NAS, 2002). Most of the phenomena which exist in open oceans can be observed in the Japan Sea. The Japan Sea, for example, has a subarctic front, many eddies of a mesoscale and the formation of deep and bottom waters due to vertical convection in winter (Senjyu, 1999; Senjyu et al., 2002, 2005; Tsunogai et al., 2003). There are various phenomena in the Japan Sea mentioned above, although their scale is smaller than those in the Pacific Ocean. Therefore, the Japan Sea can be regarded as "miniature" ocean and provides us with a natural experimental field for oceanographic studies.

The Japan Sea is easily affected by contamination compared with open oceans because of a shorter turnover time of the deep water (Watanabe et al., 1991). Since many nuclear facilities are located in the regions surrounding the Japan Sea and radioactive wastes with a total of 750 TBq was dumped into the Japan Sea by the former Union of Soviet Socialist Republics (USSR) and the Russian Federation (IAEA, 1999), the concentration of ^{129}I is important for public health of Far Eastern Countries. Moreover, the anthropogenic ^{129}I in the Japan Sea provides us information about the migration behavior of iodine in the open ocean because the Japan Sea can be thought of as 'miniature' ocean.

3. Materials and methods

3.1. Seawater sampling

Two sampling locations were selected: one is at a region off Sekine (41°23′N, 141°17′E) in Aomori Prefecture where is the boundary between the Japan Sea and the North Pacific and the other is at the Toyama Bay (37°49′N, 138°01′E: water depth 1125 m) where is the center of the Japan Sea. Surface seawater samples were collected at a region off Sekine by a submersible pump in 15 December 2006 and 20 February 2007. On 17 October 2006, deep-water samples at the Toyama Bay in the Japan Sea were collected onboard R/V Koshiji-maru using Niskin bottles (30 L) employed with the conductivity-temperature-depth (CTD) meters. The two sampling locations are shown in Fig. 1.

The seawater samples were separated into two bottles: a 100 mL polyethylene bottle for the measurement of iodine concentration and a 1000 mL polyethylene bottle for the measurement of ^{129}I by AMS. To prevent volatilization of iodine from seawater by biological processes (Miyake and Tsunogai, 1963; Muramatsu and Ohmomo, 1988), the seawater sample for the measurement of iodine concentration was frozen immediately after sampling. Although iodine in seawater samples is volatilized during storage, the iodine isotopic ratio ($^{129}I/^{127}I$) is not affected by iodine loss (Edmonds et al., 1998). Seawater samples for the measurement of the iodine isotopic ratio were brought back to an onshore laboratory without freezing.

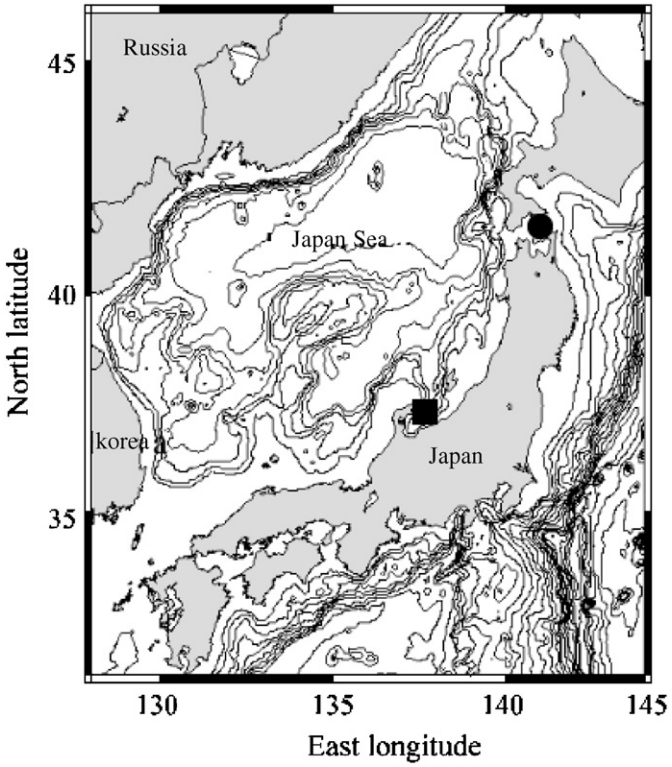

Fig. 1. Seawater sampling locations. At the sampling station in the Toyama Bay (■), seawater samples were collected from the surface to the bottom using Niskin bottles onboard by the *R/V* Koshiji-maru. At the sampling station in the off Sekine region (●), seawater samples were collected from the surface only.