



# Influence of releases of I-129 from reprocessing plants on the marine environment of the North Adriatic Sea

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## ABSTRACT

Compared to the pre-nuclear era, large amounts of  $^{129}\text{I}$  have been released to the marine environment, especially as liquid and gaseous discharges from two European reprocessing plants located at Sellafield and La Hague. Their liquid discharges influence Northern Europe and most research was conducted in the area of the North Atlantic Ocean and the Baltic Sea. In this article data on  $^{129}\text{I}$  content and  $^{129}\text{I}/^{127}\text{I}$  ratios observed in the North Adriatic Sea, which is a rather enclosed basin of the Mediterranean Sea, are presented. To the best of our knowledge no data on  $^{129}\text{I}$  in the Mediterranean Sea have previously been reported. As this area is isolated from direct liquid discharges, the main transport pathway is probably gaseous releases from reprocessing plants. Surface sea water, the marine alga *Fucus virsoides*, an iodine accumulator, and the Mediterranean mussel *Mytilus galloprovincialis* collected in 2009 and 2010, and marine sediment collected in 2005 and 2009 were analysed. The  $^{129}\text{I}/^{127}\text{I}$  isotopic ratios observed were in the range from  $0.8$  to  $3.0 \times 10^{-08}$  for seawater, from  $0.06$  to  $0.35 \times 10^{-08}$  for marine sediment, from  $0.05$  to  $0.10 \times 10^{-08}$  for *F. virsoides* and from  $0.3$  to  $0.9 \times 10^{-08}$  for *M. galloprovincialis*.

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## 1. Introduction

The only stable natural iodine isotope is  $^{127}\text{I}$  and the marine environment is the major source of iodine present on the Earth's surface, with an average concentration of  $\sim 60 \mu\text{g L}^{-1}$  iodine in seawater. Iodine is released by volatilisation from the marine environment to the atmosphere mainly as organic iodine (mostly iodomethane). Living marine organisms such as marine algae, phytoplankton and aerobic bacteria play the major roles in transformation of iodine to volatile organic forms. The iodide methylating reaction is mediated enzymatically by S-adenosyl-L-methionine as the methyl donor (Amachi et al., 2001; Amachi, 2008). The organic iodine emitted is decomposed by sunlight into inorganic iodine compounds. From the atmosphere iodine is washed out to the marine and terrestrial environment by wet (precipitation) and dry (aerosol) deposition (Muramatsu et al., 2004).

$^{129}\text{I}$  is the only natural radioactive isotope of iodine with a very long half-life ( $T_{1/2} = 1.57 \times 10^7$  y) and it is formed in nature by two processes, i.e. spallation of cosmic rays on atmospheric Xe (cosmogenic) and spontaneous fission of  $^{238}\text{U}$  (fissionogenic) (Hou, 2004). But presently, the main sources of  $^{129}\text{I}$  are nuclear fuel reprocessing plants. Although the activities of discharged radioactive liquid and gaseous effluents are of low-level and approved by the responsible

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authorities as within allowed limits,  $^{129}\text{I}$  gradually released accumulates in the environment because of its very long half life. The pre-nuclear age natural  $^{129}\text{I}/^{127}\text{I}$  isotopic ratio in the order of  $10^{-12}$  has been significantly influenced by releases of anthropogenic  $^{129}\text{I}$  (Fréchou and Calmet, 2003). The ratio of  $^{129}\text{I}/^{127}\text{I}$  in the marine environment has increased to  $10^{-11}$ – $10^{-10}$ . Anthropogenic  $^{129}\text{I}$  predominates in the biosphere and in the upper layers of the oceans.

It has been estimated that the pre-nuclear age oceans contained a total quantity of approximately 100 kg of  $^{129}\text{I}$  (Raisback and Yiou, 1999). About 5500 kg have been released to the environment since the nuclear age began and around 70000 kg of  $^{129}\text{I}$  is still pending in unprocessed nuclear fuel (Yi et al., 2011). Released  $^{129}\text{I}$  moves quickly through surface reservoirs and was found even in snow and water from the Arctic and Antarctic areas (Snyder and Fehn, 2004).  $^{129}\text{I}$  has a residence time in the troposphere of between 2 and 3 weeks (Stutz et al., 2000). In this way  $^{129}\text{I}$  is dispersed on a global scale and was found to be two orders of magnitude above natural levels even in the Southern hemisphere, where only three nuclear power stations and no reprocessing plants are located (Fehn and Snyder, 2000).

The United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR, 2008) identifies  $^3\text{H}$ ,  $^{14}\text{C}$  and  $^{129}\text{I}$  as globally dispersed radionuclides. Because of its very long half life and its accumulation in the human thyroid  $^{129}\text{I}$  is one of the most important radionuclides in long-term radiological assessment of discharges from nuclear fuel reprocessing plants. So the aim of this

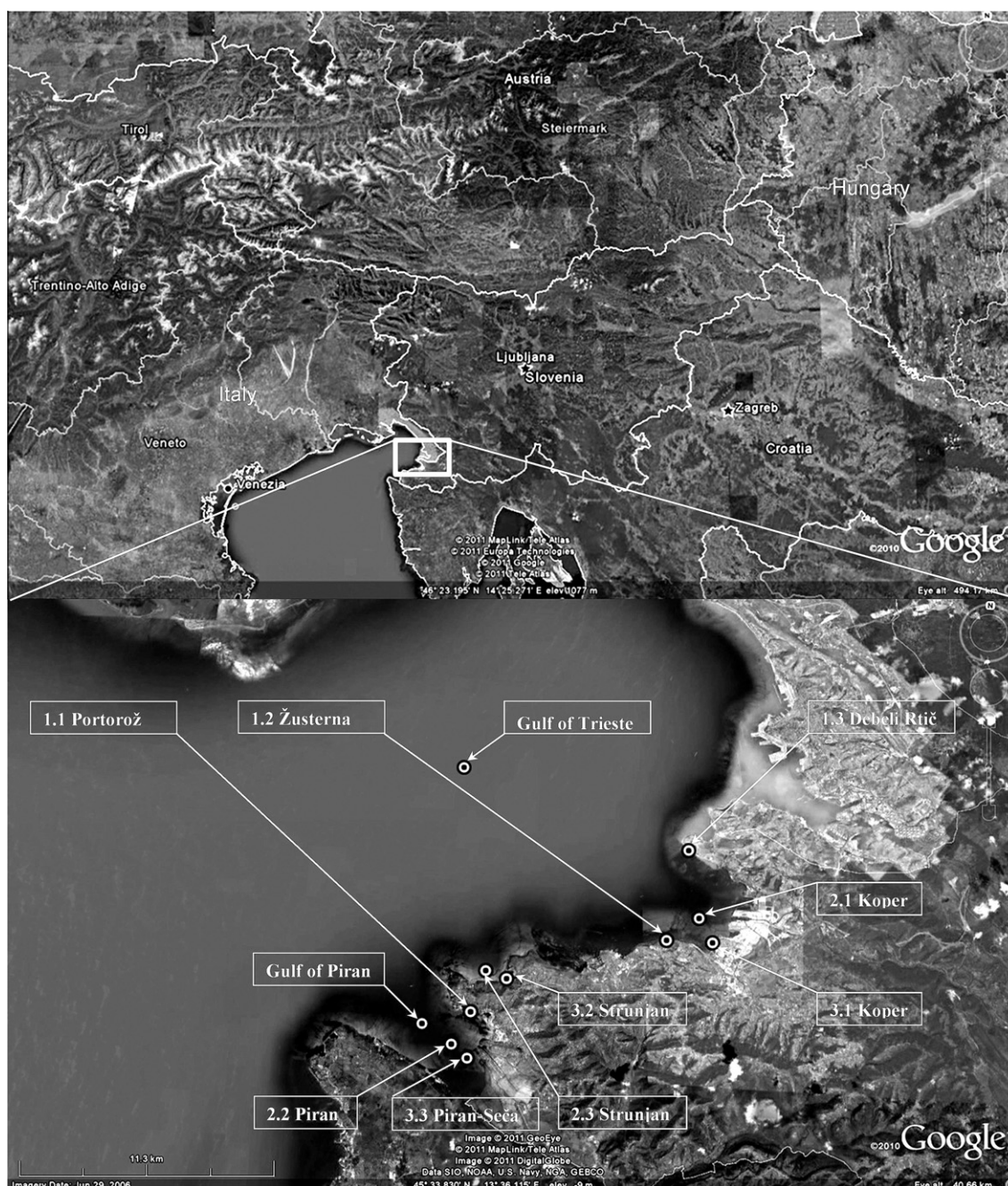
work was to investigate the  $^{129}\text{I}/^{127}\text{I}$  isotopic ratio in the marine environment of North Adriatic Sea, which lies in South East Europe, since there are no literature data available for this area or even for the Mediterranean Sea, and furthermore, it was desirable to calculate the concentration factors of iodine in some compartments of this environment.

## 2. Experimental

### 2.1. Sampling

In 2009 sampling of seawater, marine alga (*Fucus virsoides*), Mediterranean mussel (*Mytilus galloprovincialis*) and marine sediment was performed three times at the various locations shown

in Fig. 1. Simultaneous collection of the top layer of seawater and marine alga was performed at the locations of Žusterna, Portorož and Debeli Rtič. The brown alga *F. virsoides* is a species which is known to grow at the low-tide level at a depth up to 1 m and not more than 3–5 m from the shore. At each sampling location we collected a composite sample of several algae. Mediterranean mussel (*M. galloprovincialis*) samples were collected at Koper, Piran-Seča and Strunjan. One sampling of Mediterranean mussels was performed in 2010. The top layer of sediment (composite of the first 20 cm) was collected at three locations in the Bay of Piran and Koper. The sedimentation rate for western Istria, Croatia, an area very close to sampling locations, is  $2.36\text{ mm y}^{-1}$  meaning, that our sediments are a composite of last 100 y (Favre et al., 2011).



**Fig. 1.** Sampling locations; 1.1–1.3 seawater and algae: Portorož, Žusterna, Debeli Rtič; 2.1–2.3 sediment: Koper, Piran, Strunjan; 3.1–3.3 blue mussel: Koper, Strunjan, Piran-Seča.

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