Determination of $^{129}$I in conifer samples around nuclear facilities in Argentina

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An improved method has been developed that allows the determination of $^{129}$I in conifers around nuclear facilities using neutron activation analysis. Pine and cedar needle samples were collected from the surroundings of Atucha and Embalse Nuclear Power Plants, Centro Atómico Ezeiza, as well as from background areas in Argentina. $^{131}$I was used as tracer. A chemical recovery of 98–100% was obtained for pre-irradiation distillations and total recovery was 70–85%. The minimal detectable amount (MDA) was 0.01 mBq/g of sample, depending on the total recovery, natural iodine concentration, irradiation time and neutron flux. The results presented in this paper are the first published from Argentina.

Introduction

$^{129}$I is produced by spontaneous fission of natural uranium and spallation reactions of cosmic radiation with xenon in the upper atmosphere. Nuclear weapons tests and the normal operation of nuclear facilities (particularly from reprocessing plants of irradiated nuclear fuel) release small amounts of $^{129}$I in the environment, as a result of uranium fission. The combination of a long half-life ($T_{1/2} = 1.57 \times 10^{10}$ y) and a low radiation energy ($E_{\gamma} = 39.58$ keV), limits the sensitivity of the direct measurement in environmental matrices. Neutron activation analysis increases the sensitivity due to the high thermal neutron cross section of $^{129}$I (28 barn). The activity at $E_{\gamma} = 53.6$ keV of $^{130}$I generated is measured.

Iodine from nuclear power plants may appear as either elemental iodine or inorganic and organic iodine compounds adsorbed on particles and may escape from samples during the time between collection and the beginning of the process. The resinous characteristics of conifers allow the accumulation of aerosols and make them excellent matrices for $^{129}$I determination.

Several authors have used specially activated charcoal3,4 and/or quartz5–7 for adsorption/retention of iodine before irradiation. The use of a commercial charcoal for $^{129}$I adsorption and polypropylene tubes with low-sodium contents for reactor irradiation makes this method simple and suitable for routine analysis. Clean-room laboratory conditions were used during the pre-irradiation process to prevent sample contamination. By using repeated purification steps, interferences present in the environmental matrices were reduced. The main objectives were to achieve high recoveries, low detection limits, and a decrease of workers’ exposition in the post-irradiation process.

This paper also shows the concentration of $^{129}$I in environmental samples collected from the surroundings of different nuclear facilities.

Collection of conifer samples

Pine and cedar needle samples were collected from the surroundings of Atucha (CNA I) and Embalse (CNE) Nuclear Power Plants as well as around Centro Atómico Ezeiza (CAE), which are situated 100 km North, 800 km NW and 40 km SW from Buenos Aires, respectively. Background samples were collected in Buenos Aires City. Samples must be processed as soon as possible after collection in order to avoid loss of iodine.

Experimental

Collection of conifer samples

It consists of five steps as follows (Fig. 1): (1) Digestion by alkaline fusion. (2) Radiochemical purification pre-irradiation of $^{129}$I by distillation, followed by solvent extraction and adsorption on activated charcoal during a redistillation. (3) Neutron irradiation with a comparator standard, (4) Radiochemical purification post-irradiation of $^{131}$I by distillation, followed by solvent extraction, and (5) Gamma-spectrometry.

Digestion by alkaline fusion: Between 10 to 100 g of conifer needles, were placed in a nickel crucible together with twice as much KOH (s) and a (1:1) absolute ethanol—demineralized water solution enough to cover the sample. The $^{131}$I tracer was added to obtain the chemical yield. The crucible was placed on a hot plate and the temperature gradually increased up to 400°C to dry it completely. Finally, it was placed in a muffle furnace. The heating program was as follows:

Ramp 1: 10°C/min up to 450°C, this temperature was maintained for 4 hours;
Ramp 2: 10°C/min up to 600°C, this temperature was maintained for 6 hours.
This procedure was repeated as needed until a material free of organic carbonaceous particles was obtained.

Iodine in alkaline medium is present as I⁻ and in a small amount as IO⁻³. In order to keep it as iodide, K₂SO₃ must be added before the alkaline fusion.

Radiochemical purification pre-irradiation: (a) First distillation: After the alkaline fusion finished, the sample was transferred into the distillation apparatus (Fig. 2), by using deionized water. In order to avoid bromine distilling together with iodine, the alkaline solution was neutralized by slowly adding concentrated HNO₃ and it was strongly acidified with an excess of acid; then, 2 mg of KI were added as carrier. Iodine was distilled during 2 hours, adding 2–3 ml of 0.5 M KNO₂ every 30 minutes, approximately. The chemical reaction involved in the whole process is:

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2 \text{KNO}_3(aq) + 2 \text{KI}(aq) + 4 \text{HNO}_3(aq) \rightarrow 2 \text{NO}_2(g) + I_2(g) + 2 \text{H}_2\text{O}(aq) + 4 \text{KNO}_2(aq)
\]

Iodine oxosalt can be reduced by the addition of K₂SO₃. Iodine was accumulated in the collector tubes and the yield of this step was checked by measuring ¹³¹I.

The collector tubes 1 and 2 were prepared by adding 0.5 g of K₂SO₃ to 80 ml and 60 ml of 0.75 M KOH, respectively.