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Development of new low-level iodine 129 analysis method applied to the understanding of iodine's transfer mechanisms in the environment

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¹²⁹I is chronically and regularly released by fuel reprocessing plants e.g. Sellafield (UK) and ORANO La Hague (France) and also released in the atmosphere by nuclear power plants during normal operation process or in case of an accident. Iodine 129 is released by reprocessing plants not only in the atmosphere but also into the marine environment.

The objective of this study is to develop a new ¹²⁹I quantification method in order to evaluate for the first time the re-emission of iodine from the marine environment to the coast by gas or aerosol emission against direct atmospheric releases. To achieve this objective, two High Volume Sampler (HVS) are installed 2 km north of ORANO La Hague and are surrounded by the ocean except the east side. These HVS are connected to a proportional counter (LB134, Berthold). When ⁸⁵Kr is detected, the first HVS is automatically started and the collected iodine is attributed to the reprocessing plant. When ⁸⁵Kr is not detected anymore, the other HVS is started and the collected iodine is mainly attributed to reemission from marine environment. HVS contains filters to trap aerosols and coal cartridges to trap gaseous iodine. Filters and coal were selected after a study to be compatible with the analytical procedure.

In fact, a new analytical method has been developed to analyse ¹²⁹I and ¹²⁹I/¹²⁷I in these complex samples.

ICP-MS measurement following a chemical treatment is an excellent alternative to AMS for the determination of ¹²⁹I and ¹²⁹I/¹²⁷I isotopic ratio. ICP-MS technique allows reaching LOD of ~ 0,3 mBq/L for ¹²⁹I and 10⁻⁸ for ¹²⁹I/¹²⁷I (Honda et al., 2018). Difficulties during purification and measurement are related to the extreme volatility of iodine and to its multiple oxidation degrees. Spectral interferences are mainly due to the isobaric ¹²⁹Xe⁺, present as impurity in the Argon gas and to polyatomic interferences generated by ¹²⁷IH₂⁺, ¹²⁷ID⁺, ⁹⁷MoO₂⁺, ¹¹³CdO⁺, ¹¹³InO⁺, ¹¹⁵In⁺, ¹⁴N⁺ and ⁸⁹Y⁴⁰Ar⁺ (Ežerinskis et al., 2014). Non-spectral interferences are related to the matrix and memory effects.

In the presented work, the ICP-MS measurements were performed using an ICP-MS/MS (8900 Agilent®). The medium was meticulously studied to minimize spectral and non-spectral interferences. The octopole collision/reaction cell and the two quadrupole mass filters allowed minimizing polyatomic interferences. The isobaric interference due to ¹²⁹Xe was eliminated after studying different reaction gas (O₂, N₂O and CO₂) with on-mass and mass-shift detection modes.

Solid phase extraction was developed and adapted to ICP-MS measurement medium.

The new method, therefore, allows the measurement of iodine 129 either directly or after chemical treatment. The chemical treatment time has been reduced to 40 min. This step increased the test sample up to 1 L. A gain in sensitivity up to a factor 200 was observed and allowed reducing the detection limit up to 0,06 mBq/L after chemical treatment, which is 100 times lower than some current methods.

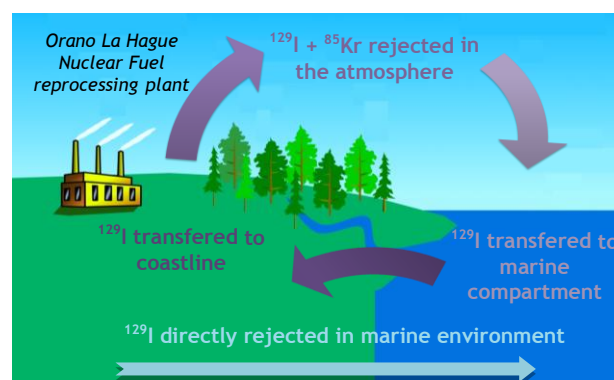


Figure 1: Iodine's transfer near to Orano La Hague

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