

# DETERMINATION OF THE $^{135}\text{Cs}/^{137}\text{Cs}$ ISOTOPIC RATIO IN ENVIRONMENTAL SAMPLES WITH LOW LEVELS OF RADIOACTIVITY USED AS A NEW TRACER FOR CAESIUM SOURCES IN THE ENVIRONMENT.

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## Abstract

Since the 1990s, the Laboratory of Environmental Radioactivity Metrology (LMRE) of the French Institute of Radiation Protection and Nuclear Safety is involved in the monitoring program of the French territory by analysing samples with very low levels of radioactivity in environmental matrices. In this context, a new methodology which allows to identify the sources of radiocaesium in the environment is being developed in the laboratory.

The isotopic signature of radioelements is a tool for characterizing different sources of radioactive contamination in the environment. For caesium (Cs), measurement by gamma spectrometry of the  $^{134}\text{Cs}/^{137}\text{Cs}$  ratio, carried out during the months following nuclear accidents of Chernobyl and Fukushima, allowed the identification of contributions from the accidental releases and the older releases sources such as the atmospheric nuclear weapons tests, the previous accidents, etc.. However, the use of this ratio is limited in time due to the short half-life of  $^{134}\text{Cs}$  ( $T_{1/2} = 2.1$  y). Thus, nowadays, 35 years after the Chernobyl accident,  $^{134}\text{Cs}$  is no longer quantifiable in France. An alternative is necessary to provide a new tracer useful for the environmental monitoring and available for the expertise of contaminated soils and sediments.

The use of the  $^{135}\text{Cs}/^{137}\text{Cs}$  ratio ( $T_{1/2}$  are  $2.3 \cdot 10^6$  y and  $30.1$  y, respectively) seems very promising in the long term but several analytical challenges exist. A major difficulty is related to the measurement of  $^{135}\text{Cs}$ . Due to the nature of its  $\beta^-$  emitted radiation, this fission product cannot be easily and accurately measured by nuclear counting. Thanks to developments over the last decade, the quantification of  $^{135}\text{Cs}$  atoms by mass spectrometry (MS) has become possible with an advanced radiochemistry and high-performance measurement (interference-free).

The current work aims at developing and validating a highly selective radiochemical protocol for the caesium purification in order to measure the  $^{135}\text{Cs}/^{137}\text{Cs}$  isotopic ratio in French environmental samples. To achieve the desired low levels of radioactivity, the first step involves completely dissolving a very large test sample of calcined sediment or soil (100 g). The caesium is then separated from the sample matrix and measurement interferences (Ba, Mo, Sb, Sn) respectively with a method using ammonium molybdophosphate (AMP) followed by two extraction resins<sup>[2,3]</sup>. Finally, the purified fraction is measured by triple quadrupole ICP-MS<sup>[1-4]</sup> under optimized reaction gas condition.

During the development of such a method, the efficiency and practicality of each step is very important. The study of each of them allowed to obtain excellent chemical yields in  $^{137}\text{Cs}$  (higher than 90 %) for 100 g of sample. In addition, in order to improve the rapidity and ease of the protocol, the compatibility of the media during the analysis was researched as well as means to reduce human interventions.

## References

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