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### Chemosphere





#### Origin and age of an ongoing radioactive contamination of soils near La hague reprocessing plant based on <sup>239+240</sup>Pu/<sup>238</sup>Pu and <sup>241</sup>Am/<sup>241</sup>Pu current ratios and <sup>90</sup>Sr and Ln(III) soil contents



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#### HIGHLIGHTS

- Determination of elevated actinides and <sup>50</sup>Sr activities near the La Hague nuclear reprocessing plant.
- Isotopic ratios show that the contamination is coming from old burnt fuel.
- The <sup>241</sup>Am-<sup>241</sup>Pu dating method yields a contamination date close to 1983.
- Elevated levels of cold light Ln (III) confirm the presence of old burnt fuel.
- The La Happe reprocessing plant operator decided to remove the contamination.

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#### ABSTRACT

Nuclear reprocessing plants are sources of environmental contamination by gaseous or liquid discharges. Numerous radionisclides are of concern, with actinides and 50Sr being the most radiotoxic. Environmental radioactivity survey programs mostly use y-spectrometry to track contaminations because yspectrometry is very cost effective and can be carried out on raw samples. On the other hand, the determination of B- or n-emitting radionuclides in environmental samples requires rather sophisticated analytical methods, and are thus dedicated to specific goals. However, measuring radionuclides such as Pa, Am, and Sr often provides more information about the presence of a current or prior contamination and on its origin, based on the isotopic composition of the samples. We found that the analysis of <sup>341</sup>Pu. 299-390 Pu. 241 Am., and 505 r of a few selected soil samples taken near the nuclear reprocessing plant of La Hague, France, revealed the presence of a previous environmental contamination originating from several incidents in La Hague site involving atmospheric transfer and leaks in flooded waste pits. The <sup>261</sup>Am. <sup>262</sup>Pu dating method indicated a contamination period prior to 1983. The presence of elevated levels of light non-radioactive lanthanides and yttrium in the soil samples confirmed the involvement of cold fuel. Our results demonstrate how long-lived actinides are likely to reveal a long-term contamination of the environment by spent fuel. Our study indicates that there is a requirement to use more sophisticated tools than y-spectrometry when surveying the environments surrounding industrial plants for nuclear power and nuclear reprocessing with a potential for the accidental release of radioactivity into the environment.

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#### 1. Introduction

Nuclear reprocessing plants are facilities that treat nuclearburnt fuel in order to: re-use some of the radionuclides for new fuel rods (Dancause et al., 2008) (e.g. MOx fuel), separate longlived radionuclides from short-lived radionuclides, and shape

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