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Empirical calibration of uranium releases in the terrestrial environment of nuclear fuel cycle facilities

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ABSTRACT

In the present paper the activity of uranium isotopes measured in plants and aerosols taken downwind of the releases of three nuclear fuel settlements was compared between them and with the activity measured at remote sites. An enhancement of ^{238}U activity as well as ^{235}U/^{238}U anomalies and ^{236}U are noticeable in wheat, grass, tree leaves and aerosols taken at the edge of nuclear fuel settlements, which show the influence of uranium chronic releases. Further plants taken at the edge of the studied sites and a few published data acquired in the same experimental conditions show that the ^{238}U activity in plants is influenced by the intensity of the U atmospheric releases. Assuming that ^{238}U in plant is proportional to the intensity of the releases, we proposed empirical relationships which allow to characterize the chronic releases on the ground. Other sources of U contamination in plants such as accidental releases and “delayed source” of uranium in soil are also discussed in the light of uranium isotopes signatures.

1. Introduction

Although the Fukushima Daiichi accident has eroded public confidence in nuclear energy in some countries, the world uranium (U) production is increasing (+7.6% since 2012) to face the overall demand of electricity that is expected to continue to grow in the next several decades to meet the needs of a growing population, particularly in developing countries (OECD, 2014). The U conversion and enrichment as well as fuel production induce U releases which reach the atmosphere and the biosphere. The contribution from U conversion and nuclear fuel fabrication in the environment surrounding the nuclear facilities is sparsely reported in the literature (Yoshida et al., 2001) and thus only a few data dealing with contamination in the air and in plants sampled in the vicinity of fuel facilities are available. Whereas the background activity of ^{238}U in the air ranges between 0.07 and 1.65 μBq m^{-3} (Masson et al., 2015), the mean values about one km downwind of a release point depend on the facility and may reach 1.4 μBq m^{-3} (ABB-ATOM, Sweden), 106 μBq m^{-3} (Springfield, UK), 40 μBq m^{-3} (Port-Hope, Canada) and 34 μBq m^{-3} (Malvési, France) (Pettersson and Holm, 1992; Al-Khayat et al., 1992; Ahier and Tracy, 1997; Masson et al., 2015). Some of those studies further report a rapid decrease of the activity in the air at further distances from the fuel facility, ultimately down to background level.

Plants sampled near fuel cycle facilities exhibit high ^{238}U activity (mean activity is 8.8 and 4.5 Bq kg^{-1} DM in cypress leaves and wheat grains, respectively) and strong variation of U activity with geometric standard deviation (GSD) reaching 6.7 (in cypress leaves) and 2.9 (in wheat grain), due to the deposition of airborne U (Pourcelot et al., 2011a, 2015a). On the other hand, plants samples taken at radiological background sites are characterized by lower activity (mean activity is 0.07 and 0.05 Bq kg^{-1} DM in grass and wheat grains, respectively) and more homogenous U activity (GSD is 2.8 and 2.9 in grass and wheat grain, respectively (Jeambrun et al., 2012). In addition, Yoshida et al. (2001) pointed out that the characterization of ^{238}U in plants and soils after a criticality accident occurring in Japan was difficult because the background level, due to chronic releases, had not been determined prior to the accident. This shows that data interpretation becomes nearly impossible when several sources of releases are present. Thus the characterization of chronic U release is crucial, especially if an accidental release superimposes on the consequences of a chronic

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emission.

On the long term, the resuspension of particles from the soil significantly contaminated by the chronic release would contribute to the contamination of the atmosphere and plants. Only few data are available dealing with this resuspension process. Thus U determined during the shutdown periods of the Malvési conversion facility (France) allow to estimate that soil resuspension contributes up to 20% of the airborne U (Masson et al., 2015).

The scope of the present study carried out in the environment of fuel facilities is to contribute to the assessment of U added in the air and in terrestrial plants by chronic discharges and hence this evaluation allows to calculate the dose to the people leaving downwind of the releases. However to reach this aim the contribution of other sources of U needs also to be determined: the contributions of the accidental releases of U, of the "delayed sources" (soils contaminated by former releases) and the contribution from the radiological background (i.e. U naturally present in the biosphere).

In this paper we present U isotope activity in plants and in the air (aerosols) sampled in the environment of three U fuel facilities: a purification and conversion to UF_4 facility (Malvési), facilities dedicated to conversion to UF_6, enrichment and defluoration (Tricastin) and a fuel rod assembly facility (Romans-sur-Isère). The characterization of the chronic releases in the atmosphere and at the ground were studying using U isotopes, which allow to quantify the excess U routinely released by each facility. We also characterize the specific isotopic ratios ($^{235}U/^{238}U$, $^{234}U/^{238}U$ and $^{236}U/^{238}U$) due to the each operation. An attempt is also made to empirically calibrate the U activity in plants with respect to the intensity of the chronic releases occurring from a facility to another. Finally the isotopic signatures observed in plants permit to distinguish and quantify sources of U different from the chronic atmospheric release, namely accidental releases and "delayed" (secondary) sources of soils contaminated by former releases of reprocessed U.

2. Studied facilities and methodology

2.1. Studied nuclear fuel facilities

The nuclear fuels facilities of Malvési, Tricastin and Romans-sur-Isère are located in Aude (SW of France), Rhône and Isère (SE of France) valleys, respectively. The purposes, the production and the yearly discharges of each site are summarized in Table 1. Malvési is mainly involved in the purification of "yellow cake" and the conversion of U to UF_4. Two steps of the conversion *i.e.* the oxidation of U to UO_3 and thereafter the reduction to UO_2 at 500 °C involve U releases to the atmosphere reaching 0.96 GBq y^{-1}. Today natural U is treated by this nuclear site and thus no isotopic anomaly (namely deviation of $^{235}U/^{238}U$ and or occurrence of man-made ^{236}U) is assumed in the present day release. However, between 1963 and 1982, reprocessed U was purified leading to the release of ^{235}U-enriched U and traces of ^{236}U and plutonium (Pourcelot et al., 2011b). Thereafter, UF_4 produced in Malvési is sent to the Tricastin facility where it is converted to UF_6 before ^{235}U enrichment. The largest part of the releases of Tricastin is induced by the conversion of UF_4 to UF_6 (0.02 GBq y^{-1}), whereas defluoration and enrichment lead to much lower releases (9.10^{-5} and 5.10^{-3} GBq y^{-1}, respectively). Other workshops from Tricastin contribute to the atmospheric releases, such as "W unit" which converts U-nitrate primarily produced in La Hague *i.e.* reprocessed U (about 1000 t y^{-1}). Thus reprocessed U released by this workshop can be distinguished from natural U by ^{236}U (Wood, 2008; Pourcelot et al., 2011b). At Tricastin, an accidental release of depleted U occurred in October 2013 which enhanced the U content in plants taken in the surroundings (Pourcelot et al., 2015b). Unfortunately the amount of U released is not known. After Tricastin, the enriched UF_6 is then transported to Romans-sur-Isère, where fuel assemblies (fuel rods) for nuclear reactors of the PWR type (Pressurized Water Reactor) are produced. At this site, the first step is the conversion of UF_6 to UO_2 powder which involves releases of enriched ^{235}U (0.0001 GBq y^{-1}).

2.2. Sampling methodology of plants and aerosols and available data

A large set of data involving plants and aerosols is available from monitoring studies carried out in the environment of the three U-fuel cycle facilities and also at sites away from the nuclear industry influence.

2.2.1. Plants

Uranium levels in plants taken near Malvési and Tricastin are reported in previous papers (Pourcelot et al., 2011a, 2015a, 2015b). In 2014–2016, additional plant samples were taken in the vicinity of Malvési (wheat grain, pine leaves and grass), Tricastin (cypress, oak and poplar trees leaves and grass and wheat grain) and Romans-sur-Isère (oak leaves, grass and wheat grain).

Those plant samples were taken in the areas potentially influenced by the U atmospheric releases, about 1000 m downwind of each facility. In the sampling area, 2–5 kg of fresh leaves, grass or wheat grain was sampled. Tree leaves were taken by hand 1–2 m above the ground. Wheat ears were collected by hand at maturity. Grass covering 1–2 m^2 was clipped with shears about 5 cm above the ground.

The activities measured in plants near U-facilities were compared with comparable samples obtained in France out of the influence of U-releases (Jeambrun et al., 2012). In this previous work, plants were taken in various geological contexts, namely granite bedrock (Vosges Mountains), leucogranites (western part of the Massif-Central), volcanic substrate (central part of the Massif-Central) and alluvial area (Rhône Valley). Thus data previously acquired by Jeambrun et al. (2012) are supposed to mirror U variations in the biosphere and will be called thereafter "background" data.

2.2.2. Aerosols

Aerosol sampling was performed downwind of the Malvési and Tricastin facilities, also about 1000 m away from the release stacks. Aerosols were sampled weekly using a home-made high-volume aerosol sampler (Masson et al., 2015). Over the whole sampling period the average regulated flow rate was about 335 $m^3.h^{-1}$

Table 1
Purpose, yearly production and atmospheric releases of three nuclear fuel facilities (2015). Data released by AREVA in the frame of the transparency and nuclear safety regulation.

Facility	Main purposes	U production (t y^{-1})	U atmospheric releases (GBq y^{-1})
Malvési	U purification and conversion to UF_4	12,000	0.96
Tricastin	Conversion to UF_6, enrichment and defluoration	12,000–13,000	0.02
Romans-sur-Isère	Fuel rod production	681	0.0001