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Environmental releases from fuel cycle facility: part 1: radionuclide resuspension vs. stack releases on ambient airborne uranium and thorium levels

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ABSTRACT

Airborne activity levels of uranium and thorium series were measured in the vicinity (1.1 km) of a uranium (UF_4) processing plant, located in Malvési, south of France. Regarding its impact on the environment, this facility is characterized by its routine atmospheric releases of uranium and by the emission of radionuclide-labelled particles from a storage pond filled with waste water or that contain dried sludge characterized by traces of plutonium and thorium (^{230}Th). This study was performed during a whole year (November 2009–November 2010) and based on weekly aerosol sampling. Thanks to ICP-MS results, it was possible to perform investigations of uranium and thorium decay product concentration in the air. The number of aerosol filters sampled (50) was sufficient to establish a relationship between airborne radionuclide variations and the wind conditions. As expected, the more the time spent in the plume, the higher the ambient levels. The respective contributions of atmospheric releases and resuspension from local soil and waste ponds on ambient dust load and uranium-bearing aerosols were estimated. Two shutdown periods dedicated to facility servicing made it possible to estimate the resuspension contribution and to specify its origin (local or regional) according to the wind direction and remote background concentration. Airborne uranium mainly comes from the emission stack and, to a minor extent (~20%), from wind resuspension of soil particles from the surrounding fields and areas devoted to waste storage. Moreover, weighed activity levels were clearly higher during operational periods than for shutdown periods.

0. Introduction

The main goal of the AREVA NC facility of Malvési (formerly Comurhex) located near the city of Narbonne in the south of France is the production of uranium tetrafluoride (UF_4) from "yellow cake"; a raw uranium material. This facility has operated non-stop since 1959. From 1963 to 1982, reprocessed uranium was purified leading to the release of uranium containing ^{236}U (Pourcelot et al., 2011a,b). Today natural uranium is treated and no isotopic anomaly is assumed in the releases. At the UF_4 facility of Malvési, considerable efforts have been dedicated to the reduction of uranium releases. One of the most recent was the installation of a scrubbing treatment at the hydrofluoridation process in 2009. This drastically reduced the amount of atmospheric emissions. As a result of the whole UF_4 industrial process and despite the reduction of the releases or filtration steps, uranium has been discharged into the atmosphere and deposited in the nearby environment. Previous studies carried out in the environment of the Malvési uranium facility detailed the industrial process and the main sources of radionuclides (Pourcelot et al., 2011a,b; Gieré et al., 2012). Pourcelot et al. (2011a) pointed out that root uptake of uranium from soil was insufficient to explain levels of uranium found in plants (crops and especially wheat grains), and suggested that atmospheric deposition had to be investigated as the main pathway involved in uranium and thorium labelling. The atmospheric deposition of uranium and thorium on plants has been treated in a closely related study by Pourcelot et al. (2015).

In order to assess the environmental impact of the uranium atmospheric releases, several monitoring strategies can be applied. Some of them are based on soil or bio-monitor samples (plant,

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crops, tree, lichen, mosses …) since they integrate atmospheric deposition and usually require low cost equipment (Bellis et al., 2001a,b; Golubev et al., 2005; Jeambrun et al., 2012). However it is not easy to discriminate between plant contamination pathways, namely atmospheric deposition or foliar capitation and root uptake. The other commonly used approach is to collect aerosol particles on air filter during more or less long periods, ranging typically 1–7 days depending on the sampler flow rate, the required detection limit and the monitoring programme.

Airborne concentration of uranium isotopes in the vicinity of a nuclear fuel fabrication facility has already been investigated (Pettersson and Holm, 1992; Meyers et al., 2014). It is well established that such industrial process impacts its nearby environment. For instance, airborne mean annual ^{238}U levels ranging 0.006–3.65 μBq m^{-3} were found decreasingly between 850 m and 5200 m from the emission stack of a nuclear fuel facility in Sweden (Pettersson and Holm, 1992) and in Canada (mean activity of 15 μBq m^{-3}) (Ahier and Tracy, 1997). However, it is not well known if long-term atmospheric releases and subsequent deposition and accumulation on soils can lead to a significant contribution to ambient airborne uranium level by resuspension compared with ongoing atmospheric releases. Conversely, airborne levels in non affected areas exhibit typical ^{238}U and ^{230}Th levels between 0.03 and 1.06 μBq m^{-3} and between 0.03 and 0.78 μBq m^{-3}, respectively (Kolb, 1989). The objective of the present work was to compare the contribution of the wind resuspension of uranium of various origins (naturally occurring uranium of crustal origin or formerly deposited from the industrial process) with that from the current stack releases. This was done by sampling at a distance of about 1.1 km by comparing the activity levels found in the air during the period of facility servicing (shutdown period without release) with a period of rated industrial release (operation period) characterized by a poor resuspension effect by wind due to wet conditions.

1. Material and methods

Aerosol sampling was performed from 6 November 2009 to 22 November 2010 at a distance of 1130 m from the main stack and at about 300–700 m from various ponds and waste storage areas. The AREVA NC facility is located in the south of France, about 3.0 km north-west to the city of Narbonne (about 50,000 inhabitants), close to the Mediterranean coast and in a rural zone (Fig. 1). The climate is typically Mediterranean with a warm and dry summer season. The sampling site (43°12′ 31″ N, 2°59′ 28″ E) was located south-east of the facility, in the 120° azimut. The prevailing wind, namely Tramontane, comes from W to NW (between 270° and 310°). It blows on average 55% of time and is characterized by high wind speeds up to 15 m s^{-1} hourly averaged. These windy prevailing conditions are favourable to wind erosion of the soil surface layer and thus resuspension of formerly deposited radionuclides. This is especially the case during summer when soil dryness is at its maximum. Annual mean precipitations are about 650 mm y^{-1} with a large variability from one year to the other (280–1175 mm y^{-1}) over the two last decades.

Aerosols were sampled weekly using a home-made high-volume sampler. Over the whole sampling period the average regulated flow rate was about 335 m^3 h^{-1} (related to standard temperature and pressure conditions). In total this corresponded to an average volume of 57,000 m^3 for each filter. Air was sampled at 1.60 m above the ground level. The sampler was equipped with a 4-layer polypropylene fibre filter (JPE 13160, Jonell Inc., USA). This filter has a minimum collection efficiency of about 95% for particles with an aerodynamic diameter of 30 nm and a collection efficiency of at least 99% for particles with an aerodynamic diameter greater than 0.15 μm. Aerosol filters were thermally pressed into a pellet before their measurement by gamma spectrometry on low-background high-purity Germanium (HPGe) γ-detectors.

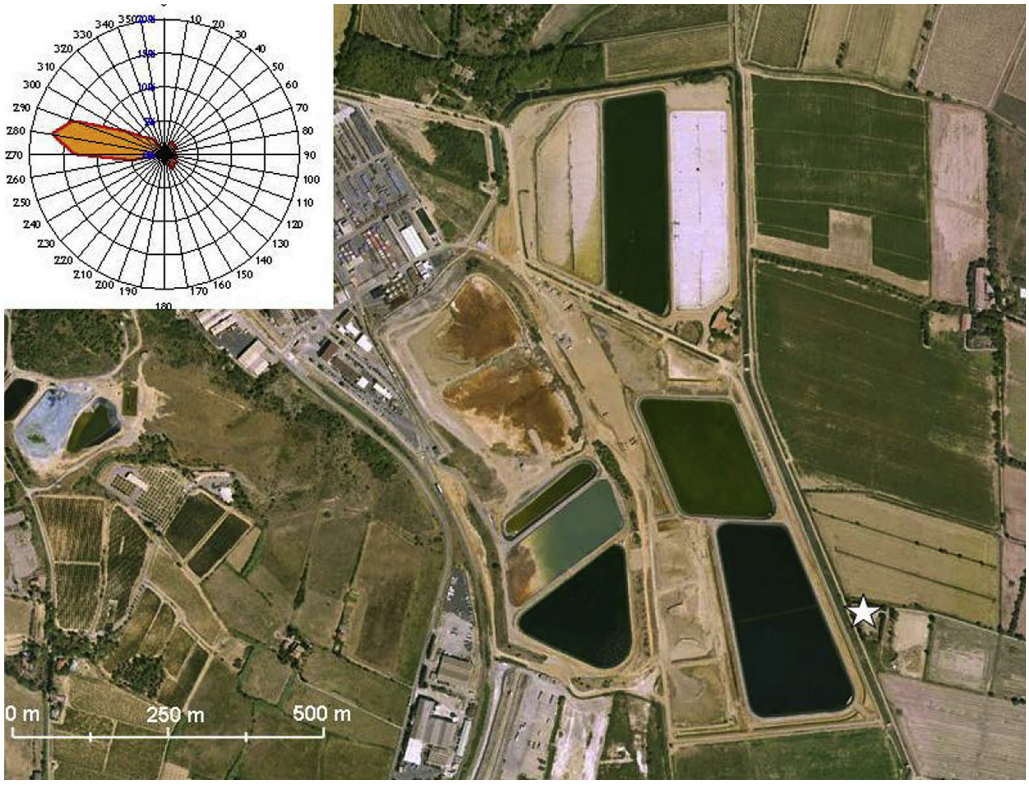

Fig. 1. Aerial view of the AREVA NC facility in Malvési, wind rose and sampling location (white star).