IRSN
INSTITUT
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How are radioactive deposits redistributed within catchment basins in post-accidental situations?

Lessons learned from the Chernobyl and Fukushima accidents

Following the Chernobyl and Fukushima accidents, radioactive releases into the atmosphere led to heterogeneous deposits on vast mainland surfaces including forests, agricultural land and residential areas. In the contaminated zones, the downstream redistribution of radiocaesiums[1] associated with the initial deposits is governed by hydrological processes such as surface runoff or transport into waterways. Knowledge about the dynamics of flows of radionuclides associated with these processes is essential in order to optimise post-accident management strategy, in particular for the use of aquatic resources by human populations.

The leaching of the catchment basins governs the redistribution of the radionuclides deposited on the soils following a nuclear accident; in both Chernobyl and Fukushima, this process annually remobilises less than 1% of the stock of deposited radiocaesiums. The long-term radioactive contamination of the catchment basins (mainland surfaces - soils, lakes and rivers - from which all of the water converges towards the same outlet) originates from the atmospheric deposits of radioactive contaminants released during the Chernobyl and Fukushima nuclear accidents, like those from the former atmospheric testing of nuclear weapons The leaching of these systems - i.e. the entrainment of the radionuclides by the water running on the soil surface - is a persistent dispersion process of radionuclides in the environment and over large distances, sometimes beyond the initially contaminated and/or evacuated territories (Gulin *et al*., 2013). This hydrological process induces both a redistribution of the contamination in the catchment basins (Walling *et al*., 1999; Khanbilvardi *et al*., 1999; van der Perk et Slavik, 2002) and its export downstream of the hydrographic networks (*e.g.,* Menzel, 1960; Yamagata, 1963) to the sea (Kakehi *et al*., 2016). Leaching is a process that, through the remobilisation of the radionuclides deposited after the accident, influences the radiological impact on exposed human populations based on the uses of environmental resources from the aquatic ecosystems situated downstream of the leached territories (waterways, lakes, dams used for irrigation and the supply of drinking water, fishing). This leaching does not however induce, over the long term, a significant decontamination of the contaminated territories: as observations made at Chernobyl have shown, annually, leaching does not entrain more than 1 percent of the radionuclides present on the soils (Borzilov *et al*., 1988; Khanbilvardi *et al*., 1999). This order of magnitude is identical in the case of Fukushima (e.g., Ueda *et al*., 2013).

The flows of radiocaesiums remobilised from the soils of the catchment basins are transported by water and particles; this is the result of complex chemical, hydrological and erosive processes that are expressed in two phases that differ both kinetically and with regard to the effectiveness of flows exported towards the outlet. During leaching, radionuclides are transported by two vectors: water, if they are in dissolved form (via the runoff process) and particles if they are fixed to suspended matter (erosion), which leads to a distinction between "liquid" leaching and "solid" leaching (Bulgakov *et al*., 1991). Leaching is a downstream radionuclide

1 These radiocaesiums are caesium-134 and caesium-137.

transfer process from the catchment basins that remains difficult to assess, particularly due to the complexity of runoff and hydric erosion (Figure 1). This leaching depends notably on the types of rainfall (intensity, volume of runoff water, time intervals between rainfall occurrences, etc.) and leached surfaces (texture and porosity of the soil, type of plant cover, slope, etc.), whose characteristics can often be heterogeneous within a catchment basin and variable over time.

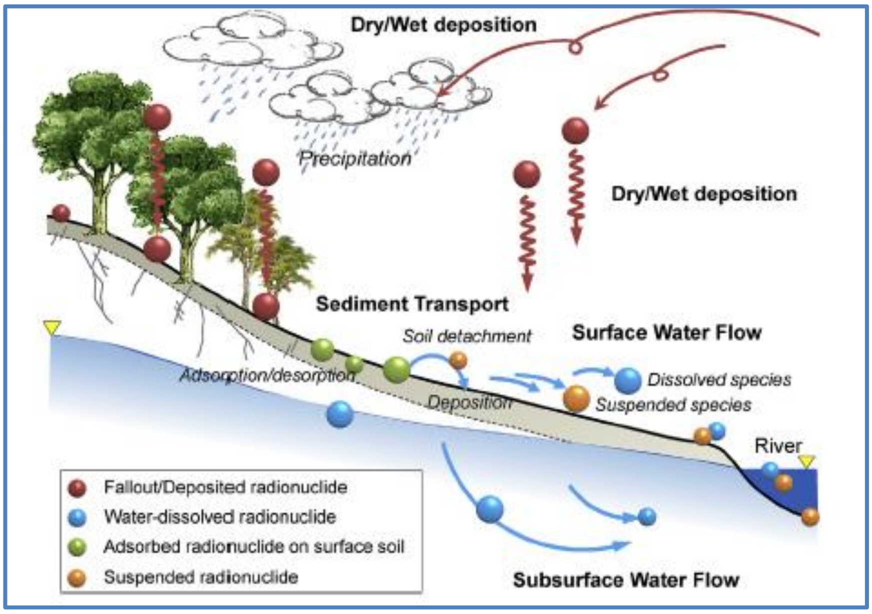

Figure 1 - Schematic representation of the redistribution processes of radionuclides deposited within a catchment basin following a nuclear accident (from Mori *et al.*, 2015).

We roughly know the main phases and orders of magnitude of this leaching following an accidental atmospheric deposit, notably for caesium-137. For this radionuclide, on a monthly or even annual scale, there is the rapid leaching phase during the first weeks after the accident, where flows of caesium-137 are high and export around 1% of the initial deposit onto the catchment basin; and the slow leaching phase, where exported flows are much lower, at around 0.1% per year of the stock still present in the catchment basins. The exported flows however remain detectable several decades after the deposit, as has been observed with the overall fallout of caesium-137 released during former aerial nuclear tests (Helton *et al.*, 1985).

The work carried out by IRSN as part of the AMORAD project[2] for the Nitta coastal river in the Fukushima area (Delmas *et al.*, 2016) has enabled an estimation of a radiocaesium export rate during the first year following the accident of 0.12 to 0.8% in the Nitta basin, which is lower than the values proposed by Kinouchi *et al.* (2015) and Ueda *et al.* (2013) for small catchment basins in the study zone. This can be explained by the difference in surface area of the catchment basins (scale effect) as well as by the presence of a dam in the Nitta basin that limits the entrainment of radionuclides towards the basin outlet.

At timescales of under a year, leaching is very erratic, in particular under the influence of climatic events (*e.g.* rainfall, typhoons, snow melt) (Ueda *et al.*, 2013). Moreover, the radionuclides are mobilised in space in a heterogeneous manner under the influence of variable runoff conditions (*e.g.* diffuse or channel runoff on slopes, concentrated runoff in the hydrographic network), and their course includes transitory storage areas (*e.g.* floor sediments in lakes/dams, tidemarks on banks).

[2] The AMORAD project is co-funded by the ANR RSNR ("Improvement of dispersion and transfer model prediction and evaluation of the environmental impact of radionuclides ").

The difficulty in transposing the research findings acquired after the Chernobyl accident to estimate exported flows in the case of Fukushima has different origins. The main differences between the two situations are linked to the availability for leaching of the radionuclides deposited on the soils, and to differences in topography and weather patterns. The outcome is that the leaching of radiocaesiums in Fukushima is mainly solid (vs. liquid in Chernobyl) and that in both cases floods explain the majority of annual exports towards the marine environment. In Fukushima, the major significant radionuclides to take into consideration for the medium- and long-term management of radiation protection are the radiocaesiums deposited in the form of aerosols, whereas the spectrum of radionuclides deposited following the Chernobyl accident is much wider with, notably, isotopes of strontium, caesium, americium, plutonium and curium (Matsunaga *et al.*, 1998). In the Chernobyl exclusion zone, hot fuel particles released during the explosion of the reactor were also dispersed. In the vicinity of the Chernobyl power plant, contamination is therefore mainly in the form of particles of insoluble fuels containing, notably, radiocaesiums and radiostrontium which are progressively released during their alteration and disintegration (Konoplev *et al.*, 1992). At Fukushima, caesium is considered to be mainly deposited in the form of soluble sub-micronic aerosols (Kaneyasu *et al.*, 2012), but the minority presence of insoluble forms has also been confirmed (Tanaka *et al.*, 2013; Adachi *et al.*, 2013).

Despite the types of deposits in Fukushima being, *a priori,* more sensitive to "liquid" leaching than in Chernobyl, at Fukushima the radiocaesiums are mainly transported in particulate form during leaching (Sakaguchi *et al.*, 2015; Konoplev *et al.*, 2016; Tanaka *et al.*, 2013; Niimura *et al.*, 2015), whereas they are largely carried in soluble form in Belarus and in the Ukraine (Gorbachova, 2015). Numerous explanations have been put forward:

- suspended matter at Fukushima contains more clay minerals than at Chernobyl and has a greater affinity to caesium; for example, the solid-liquid distribution coefficient of radiocaesiums[3] in Fukushima rivers (ca. 10^5 to 10^6 L/kg) are one to two orders of magnitude higher than those in the Chernobyl area (ca. 10^4 L/kg) (Eyrolle-Boyer *et al.*, 2016; Konoplev *et al.*, 2016);
- the presence of humic acids in Ukrainian waters inhibits the sorption of caesium on suspended matter (Suga *et al.*, 2014);

On a topographical level, the contaminated catchment basins in Fukushima are smaller, steep and are subject to greater climate extremes (Konoplev *et al.*, 2016). The Chernobyl exclusion zone is in the gently-sloping flood plain of a large river, the Dniepr, where rainfall is around 500 mm/year and in the form of snow on frozen soils several months per year. The catchment basins in the Fukushima prefecture are drained by small coastal rivers with many constructions (weirs, dams, irrigation channels) less than 100-km long, and are subject to rainfall of around 1,200 mm/year, alternating between tropical cyclones from July to October and snow melt in the spring. Despite these contrasting meteorological patterns in the Ukraine and Japan, in both cases flood events are behind most of the annual contamination exportation: these floods occur especially during snow melt in the Ukraine (Borzilov *et al.*, 1988), whilst they also occur as a result of typhoons in Japan, where the rainfall is much more erosive (Ueda *et al.*, 2013). These events generate 80 to 90% of annual exports to the marine environment in the Fukushima zone.

The initial work by the IRSN as part of the AMORAD project (Delmas *et al.*, 2016) shows that the entrainment coefficients (proportion of exported soil contamination for 1 mm of runoff water per m^2 of soil or 1 g of suspended matter exported per m^2 of soil) decrease over time in the study zone following the Fukushima accident, with maximum values after typhoons. These liquid entrainment coefficients are lower, by one or two orders of magnitude, in the Fukushima context than in Chernobyl (Konoplev *et al.*, 2016), whilst the solid entrainment coefficients do not seem to differ between the catchment basins of these two damaged zones. This comparison confirms the major role of particles in the leaching of caesium in the Japanese context. The specificity of the Japanese

[3] The solid-liquid distribution coefficient of a radionuclide is the equilibrium ratio between the radionuclide concentration in particles (Bq/kg) and that in water (Bq/L)

context is in all likelihood the result of solid particles with greater caesium affinity, on the one hand and, on the other hand, more intense erosion processes.

Finally, a study carried out by Yoshimura *et al.* (2015) on 30 catchment basins in the prefectures of Fukushima and Miyagi in December 2012 showed significant correlations between radiocaesium concentrations in dissolved form or associated with particles in the waterways, and the stock of radioactive deposits characterising the catchment basin where the waterway is located. These findings were recently confirmed by studies carried out by the IRSN (Eyrolle-Boyer et al., 2016; figure 2). This point underlines the relevance of the scale of the catchment basin in order to assess the flows exported towards the marine environment. At this observational scale, Mori *et al.* (2015) estimated, on the basis of models validated by measurements *in situ*, that over 90% of the radiocaesiums deposited on the catchment basin remain present three years after the accident. Assuming that the redistribution of caesium-137 continues in an identical way after this three-year period, the time taken to export 50% of the initial stock deposited on the catchment basin has been estimated at around 18 years (Mori *et al.*, 2015). This estimation takes into account the flows exported by liquid and solid runoff as well as radioactive decay.

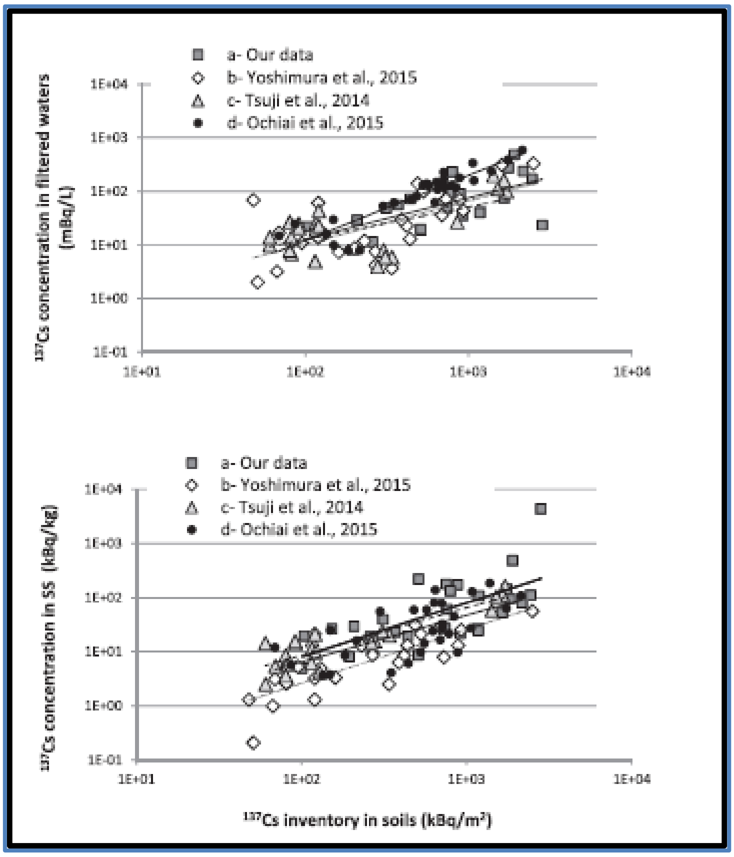

Figure 2- Relationship between concentrations in particulate or dissolved radiocaesiums and the stock of radioactive deposits (Cs^{137}) at the catchment basin level. The study focuses on the sites sampled between September 2012 and February 2013 (from Eyrolle-Boyer *et al.*, 2016).

The strategy applied to the management of aquatic environments in Chernobyl and Fukushima is different. Countermeasures have mainly concerned aquatic environments in Chernobyl, where the river Dniepr and its cascade of reservoirs downstream to the Black Sea are the main Ukrainian water supply source (Voitsekhovitch *et al.*, 1997). Sediment traps were built in 1986-1987 but their effectiveness proved limited as they did not retain fine particles - which are the most contaminated. The contaminated areas in the exclusion zone in the vicinity of the power plant (on the Prypiat river) were flooded in 1988 and 1991, and constitute the main source of contamination of the Prypiat river. Starting from studies based on the modelling of transfers in the zone, dykes were built on the left bank in 1992-1993 and their effectiveness was proven during the floods of summer 1993, the logjam of winter 1994 and the 1999 rise in water levels. Dykes were also added to the right bank in 1999-2002, but development work has since been suspended. Further downstream, the Dniepr is a river regulated by a series of dams, whose management has been optimised in order to limit the transfer of radionuclides to the Black Sea.

In Fukushima, even though a change in dam management methods is under consideration (Yamada *et al.*, 2015), countermeasures remain massively focused on the decontamination of the territories. In agricultural areas, and around houses, the removal of the upper soil layers and their replacement by non-contaminated soil aim for a reduction in the ambient dose rate and the rehabilitation over time of certain zones evacuated at the time of the accident. The impact of the mass decontamination operations on the kinetics and intensity of flows exported by the leaching of catchment basins is as yet unknown. At this stage, it is estimated that the flows of caesium-137 entrained towards the ocean by the leaching of the stocks deposited in the area 100 km around the Fukushima-Daiichi power plant (overall flow of 8.4 TBq/year of caesium-137 spread over the maritime facade, which stretches towards the north from the Fukushima-Daiichi power plant to the mouth of the Abukuma), are of the same order of magnitude as the direct releases from the damaged Fukushima-Daiichi plant (estimated at 17 TBq between June 2011 and September 2012, and at 24 TBq during a release that took place on 21 August 2013 (Yamashiki *et al.*, 2014)).

In addition to agricultural, urban and forest areas, Japan has also integrated river and lake management into its recovery plan for the territories impacted by the fallouts of the Fukushima accident. The driving concept is based on the fact that the presence of a water column above the contaminated sediments ensures a protective effect that, very effectively, reduces the dose rate associated with external exposure of human populations to contaminated sediments. Decontamination therefore only takes place in cases where this protective effect is insufficient in order to limit the increase in the ambient dose rate (e.g. lowering of the water level, drying out). This is the case, for example, for public spaces developed for various human activities established on the banks of waterways, or for reservoirs situated in residential areas or parks. Continued monitoring of the level of radioactivity in aquatic environments as well as research and development projects in order to understand and quantify the fate of radionuclides at a global scale with regard to catchment basins are in progress (Ministry of the Environment, Japan, 2016). IRSN research, as part of the AMORAD project aimed at quantifying and modelling the remobilisation processes of radionuclides through the leaching of catchment basins at different temporal scales (from one episode of rainfall to several years) will contribute to the consolidation of elements of post-accidental management of aquatic environments.

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