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ARTICLE

Origins and trend of radionuclides within the lower Rhône River over the last decades

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Abstract – Geological and climatic diversity leads to significant spatial variability of naturally occurring radioactivity levels, whether in soils, sediments or natural waters. The activity level of Rhône sediments is estimated at 1450 Bq/kg, between the levels observed in the Loire (1925 Bq/kg) and Seine (730 Bq/kg). The largest amounts of radioactive effluent from nuclear facilities concern tritium, for which activity levels are currently 1000 times higher than the sum of artificial gamma emitters discharged. The proportions of naturally occurring ^{14}C and ^{3}H in the lower reaches of the River Rhône are estimated at 50 to 70% and <5%, respectively, with the remaining amount coming from nuclear facilities. Long-term records from River Rhône monitoring show that the level of radioactive contamination from artificial sources declined sharply starting in the early 90s, with the level divided by 10 to 100 depending on the element. Radioactivity of natural origin remained unchanged as expected.

Keywords: Rhone river / radionuclides / chronicles / suspended particles / filtered waters

1 Introduction

The 19 nuclear power plants (NPPs) in France are licensed to discharge low-level, radioactive liquid effluent to aquatic systems, subject to regulations and inspections. The systems concerned are, in descending order of total installed power output: the Channel (15 800 MW), the Loire and the Vienne (14 500 MW), the Rhône (13 400 MW), the Garonne (6 200 MW), the Moselle (5200 MW), the Meuse (2900 MW), the Seine (2600 MW) and the Rhine (1800 MW). In addition to NPPs, the Marcoule spent fuel reprocessing plant, downstream of Tricastin, which has been in the decommissioning phase since 1997, has also discharged waste into the lower reaches of the Rhône since 1958. Over the last two decades, some 20% of gamma-emitting liquid effluent from the 19 NPPs in France has been discharged into the Channel. Besides the marine environment, more than 60% of NPP effluent has been discharged into the Rhône (22%) and the Loire (39%). Only 1% of effluent was discharged into the Seine. From the report, it emerges that the Rhône received nearly 65% of gamma-emitting liquid effluent discharged into the continental aquatic environment by nuclear facilities of all categories. Almost 85% of this quantity was due to the Marcoule reprocessing centre (Eyrolle, 2009).

The radioactivity of water in the River Rhône has been monitored since the 1960s. Monitoring developed further in the 1970s, in particular with the introduction of systematic gamma spectrometry and tritium analysis methods. Monitoring focuses on two targets: dissolved elements and the particulate fraction, in the first case by analysing filtered water, and in the second through the analysis of settling sludge or suspended matter. One of the sampling stations on the Rhône, located in Vallabrègues, some sixty kilometres from the mouth of the river, was operated initially by the SCPRI[1], then by the OPRI[2] and now by the IRSN. Sampling is carried out there using a "hydro-collector" designed to collect water continuously from the Rhône to obtain monthly samples of filtered water and suspended matter. For more than a decade, the SORA monitoring station in Arles has been used to collect frequent water samples from the Rhône to determine the activity levels of gamma-, beta- and alpha-emitting radionuclides. The Arles monitoring station is located 3.5 km downstream of the diffluence with the Petit Rhône and 45 km upstream of the river mouth of the Grand Rhône, that registers 85% to 90% of the liquid and solid flows of the River Rhône. By collecting samples of large volumes of water, partially dependent on the flow of passing water and implementing high-performance metrology, the SORA monitoring station allows the detection of artificial radionuclides, now found in trace concentrations, particularly in filtered water from the Rhône (Eyrolle *et al.*, 2010).

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This article shows trends in activity levels due to naturally occurring and artificial radionuclides observed over the last few decades in the lower reaches of the River Rhône.

2 Results and discussion

2.1 Origins and trends of radionuclides in the lower Rhône River over the last few decades

2.1.1 Naturally occurring radionuclides

Most of the naturally occurring radionuclides in the Rhône are drained from soil in the river's drainage basin. These are very long-lived elements which have always been found on Earth: ^{238}U, ^{235}U, ^{232}Th and ^{40}K for the most part. The first three decay to produce about thirty decay products or daughter radionuclides with highly variable half-lives, including other isotopes of uranium (^{234}U) and thorium (^{232}Th, ^{230}Th and ^{228}Th), radium isotopes (^{226}Ra, ^{228}Ra and ^{224}Ra), and radioactive isotopes of lead (^{210}Pb in particular). Naturally occurring radionuclides are also formed continuously in the atmosphere as a result of cosmic radiation. These include tritium (^{3}H), carbon-14 (^{14}C) and beryllium-7 (^{7}Be) in particular. These cosmogenic radionuclides are deposited on the surface of the soil, especially by precipitation, and are then drained into the river from soil in the drainage basin. Owing to geological and climatic diversity throughout France, and even at a regional level, natural radioactivity is subject to significant spatial variability and is highly dependent on geology and environmental conditions, whether in soils, sediments or natural water (Picat *et al.*, 2002; Porcelli and Swarzenski, 2003; Pourcelot and Vassas, 2005; Le Roux, 2007). The decay chain of ^{238}U and ^{232}Th, respectively, includes 14 and 10 decay products, including ^{226}Ra and ^{210}Po in the first case and ^{228}Ac in the second. As different decay products are never exhaustively quantified at the same time, the assessment of the natural radioactivity of water or sediment is based on the assumption that two elements measured in the same decay chain are at radioactive equilibrium. In this case, the natural radioactivity of water can be approximated by the following equation (1):

$$\begin{aligned}\text{Natural radioactivity} &\cong [^{40}\text{K}] + (14[^{238}\text{U}] + 10[^{232}\text{Th}]) \\ &\cong [^{40}\text{K}] + (14[^{234}\text{Pa}] + 10[^{228}\text{Ac}]). \quad (1)\end{aligned}$$

This calculation was used to estimate the activity of naturally occurring radionuclides in sediments in the main rivers in France. The results of these calculations show that Rhône sediments exhibit natural radioactivity levels in the region of 1450 Bq/kg, which is between the values observed in the Loire (1925 Bq/kg) and Seine (730 Bq/kg) (Eyrolle *et al.*, 2008). For comparison, the natural radioactivity of sediments from the Têt, a coastal river in the department of Pyrénées Orientales, west of the Hérault, is between 2000 and 2500 Bq/kg. These higher natural radioactivity values can be explained by high ^{40}K concentrations, which are generally an indication of an anthropogenic contribution related to the use of agricultural fertilisers (Ferrand, 2010; Ferrand *et al.*, 2012).

2.1.2 Artificial radionuclides

Artificial radionuclides have been released to the environment since the mid-twentieth century through the use of nuclear energy, first for military purposes, then in industry. The artificial radionuclides found in the Rhône today have several origins.

2.1.2.1 Persistence from atmospheric fallout

Total atmospheric fallout from nuclear tests performed between 1945 and 1980 combined with that from the Chernobyl accident has had an impact on environmental radioactivity. For example, the nuclear surface tests carried out between 1945 and 1980 led to an increase in atmospheric tritium concentrations by three orders of magnitude (IAEA, 2012) and those of ^{14}C by about a factor of 2.

Although most of the short- and medium-lived elements from this fallout have now disappeared as a result of the decay process, residual activity can still be observed in the soil in the Rhône basin, mainly due to ^{137}Cs, ^{90}Sr and transuranium long-lived elements (^{238}Pu, $^{239+240}$Pu and ^{241}Am) (Perkins and Thomas, 1980; Duffa, 2001; Renaud *et al.*, 2004). Soil erosion and draining processes lead to the transfer of these artificial radionuclides to river water in dissolved form and/or bound to solid particles. Since the early 1990s and the sharp decline in the discharge of liquid effluent from nuclear facilities along the Rhône, the presence of artificial radionuclides in the river via its drainage basin has represented a considerable and, in some cases, predominant source term. This is the case in particular of ^{137}Cs and plutonium isotopes when the river is in flood (Eyrolle *et al.*, 2006; Rolland *et al.*, 2006). It is also the case of ^{14}C, where terrigenous contributions would seem to account for 50 to 70% of the annual average activity recorded in the lower Rhône (Eyrolle-Boyer *et al.*, 2012). It is estimated, however, that less than 5% of the annual average free tritium (HTO) activity in the lower Rhône is attributable to the drainage basin, with effluent from the nuclear industry accounting for most of this activity (Eyrolle-Boyer *et al.*, 2013).

2.1.2.2 Liquid effluent from nuclear facilities

The main source terms in the Rhône valley are the Marcoule industrial site and the Bugey, Saint-Alban, Cruas and Tricastin nuclear power plants and, to a lesser extent, the Pierrelatte site and the Creys-Malville site, which is currently undergoing deconstruction.

Tritium represented the greatest amount of effluent discharged by nuclear facilities, with a total activity of 287 TBq in 2010, all types of facilities considered. Excluding ^{14}C, the activity of the other radionuclides discharged into the Rhône (for the most part ^{58}Co, ^{60}Co, ^{63}Ni, ^{90}Sr, ^{106}Ru, ^{125}Sb, ^{110m}Ag and ^{137}Cs) totalled 125 GBq in 2010, which is about 2000 times less than the activity of tritium discharge alone; ^{14}C releases from nuclear industries (excluding Marcoule discharges) being of the same order of magnitude (149 GBq in 2010) (Fig. 1).

While most of the tritium observed came from nuclear power plants, the Marcoule site was responsible for 95% of activity due to other radionuclides.

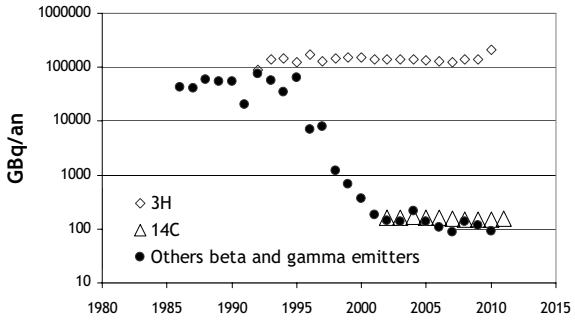

Fig. 1. Records of activity levels for tritium, ^{14}C and other radionuclides (in GBq/year) discharged into the River Rhône.

A considerable decline in the quantities of radionuclides discharged to the river has been observed over the last two decades (Fig. 2). The most significant downturns were observed starting in 1990, when a new plant was opened to treat liquid effluent from the Marcoule site and again in 1997, when spent fuel reprocessing came to an end in Marcoule.

The isotopic composition of effluent has considerably changed over time, especially effluent from Marcoule and, to a lesser extent, nuclear power plants. This change is mainly due to the quality of reprocessed fuel and waste treatment methods. Between 1975 and 1990, ^{106}Ru was by far the most abundant gamma-emitting radionuclide contained in liquid effluent from the Marcoule site. At present, ^{90}Sr, ^{137}Cs and ^{60}Co account for 90% of the activity discharged by the site (Eyrolle and Renaud, 2012; Eyrolle-Boyer *et al.*, 2012). After tritium, which represented more than 95% of radioactive effluent, the principal radionuclides discharged by nuclear power plants were ^{58}Co (22%), ^{110m}Ag and ^{60}Co (21%), and ^{63}Ni (17%).

2.1.2.3 Hospital waste

A number of medical centres using nuclear sources for diagnosis and treatment purposes are located along the Rhône and some of its tributaries. Discharge from these centres contains very short-lived radionuclides, of which only iodine-131 (^{131}I) is regularly measured in the Rhône.

2.1.2.4 Recent significant events – atmospheric fallout in France from the Fukushima accident

The accident at the Fukushima NPP in Japan led to an extremely small amount of fallout in France, observed mostly between mid-March and early May 2011, and equivalent to about one-thousandth of the fallout in France from the Chernobyl accident. The analysis of the impact of the Fukushima accident in France based on the results of reinforced environmental radioactivity monitoring was summarised in the IRSN report IRSN/DEI/2011-01 (Collectif DEI, 2011). No significant observations could be made of the impact of the Fukushima accident in French rivers, either because the related activity levels were too low, or because they were masked by activity due to discharge from nuclear facilities (^{134}Cs and ^{137}Cs), by residual activity from the Chernobyl accident (^{137}Cs), or by hospital waste, as in the case of ^{131}I, which was the most abundant radionuclide observed in the air and in food after the accident. By the end of spring 2011, any ^{131}I resulting from the Fukushima accident had completely disappeared from all compartments of the environment in France, owing to its short radioactive half-life of 8 days. It is worth noting that during annual radioecological monitoring studies conducted in 2011 and 2012, ^{134}Cs, which had disappeared from the environment for several years, was nevertheless detected in several samples of bryophytes, which are aquatic mosses considered as good bioindicators, at levels very close to quantification thresholds (Claval *et al.*, 2012), probably attesting to some inputs from the Fukushima accident into French aquatic systems.

2.2 Activity levels measured over the last few decades

Environmental monitoring records show that while natural radioactivity has obviously remained stable over time, contamination levels in the River Rhône began to decline sharply in the early 1990s, being divided by 10 to 100 according to the radionuclide concerned. This can be explained by the reduced amounts of liquid effluent from the Marcoule site, as demonstrated in Figure 2.

Whatever their origin, radionuclides were found in the dissolved phase (filtered water) or the particulate phase (suspended matter), depending mainly on their chemical properties; in other words, on their ability to bind to solid suspended particles or be deposited as sediment (Eyrolle and Charmasson, 2001, 2004; Filella *et al.*, 2002; Rolland *et al.*, 2006). Monitoring records revealed changes in activity levels in both these phases.

2.2.1 Activity levels measured in filtered water

2.2.1.1 Naturally occurring radionuclides

^{40}K and the various elements in the ^{238}U and ^{232}Th decay chains, such as ^{234}Pa and ^{228}Ac, accounted for most of the natural radioactivity in the River Rhône (Fig. 3). ^{7}Be, a short-lived (53 days) cosmogenic radionuclide was observed at activity concentrations that were significantly lower and more variable than those of various telluric radionuclides. The activity of this radionuclide in water largely depends on the age of the water mass in transit and on precipitation over the drainage basin. The activity levels of ^{22}Na, another cosmogenic radionuclide, were an order of magnitude lower.

As expected, the activity levels of naturally occurring radionuclides measured in filtered water from the lower Rhône were stable over time (on a human scale), and consequently the same as values recorded in previous years. This was the case for naturally occurring radionuclides routinely measured by gamma spectrometry, such as those shown in Figure 3. It was also the case, however, for naturally occurring isotopes that are not investigated on a routine basis, but that can be quantified by isotopic analysis. These include in particular ^{226}Ra, ^{228}Ra, ^{230}Th, ^{232}Th and ^{234}U.

Natural radioactivity measured in filtered water throughout France was in the region of a few hundred mBq/L. On the whole, this natural radioactivity was distributed almost equally among ^{40}K and the ^{238}U and ^{232}Th decay chains, both in the

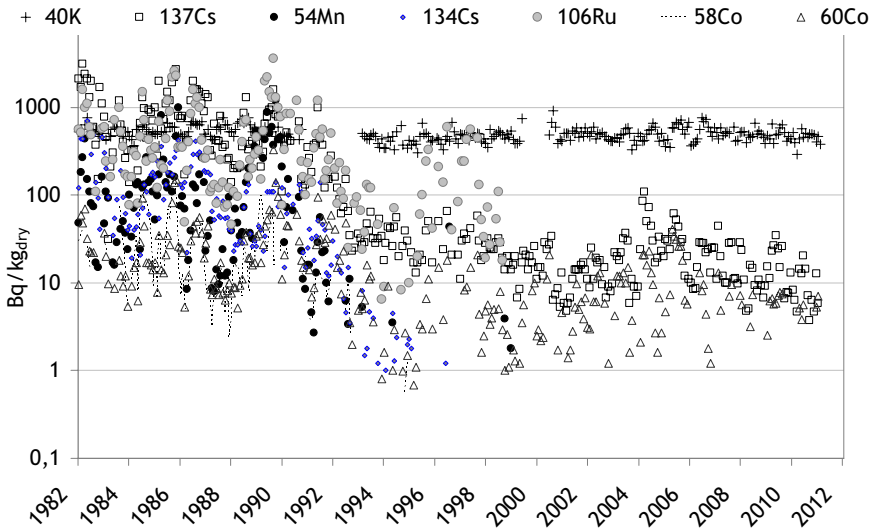

Fig. 2. Records of activity levels measured in suspended matter in the lower Rhône over the last few decades (Vallabrègues monitoring station). Since the beginning of the 21st century, only the artificial radionuclides ^{137}Cs and ^{60}Co have been routinely detected by gamma spectrometry at this station.

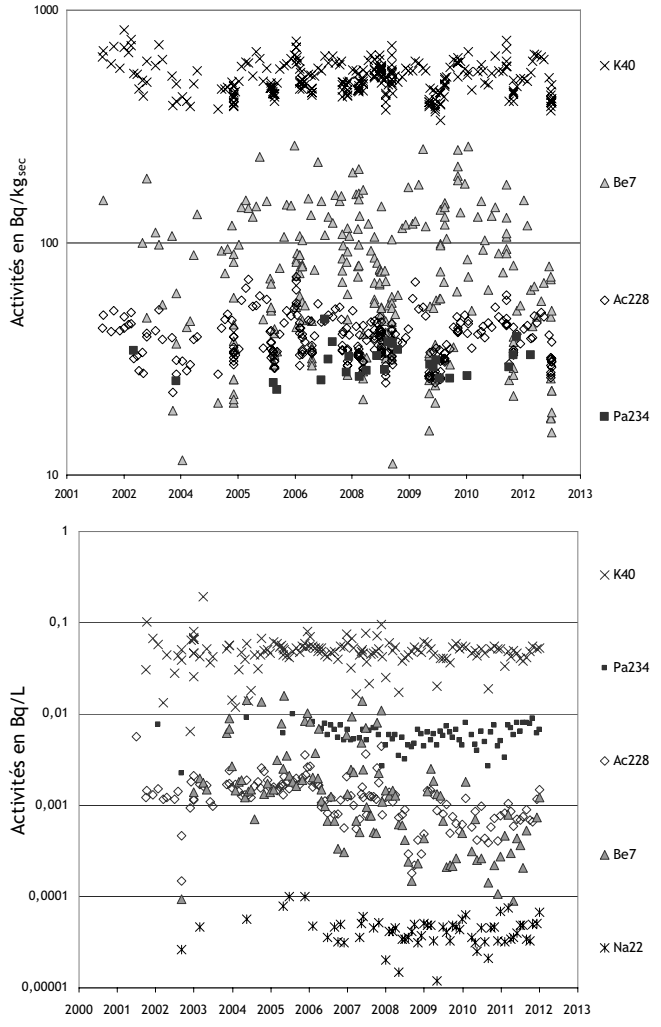

Fig. 3. Record of activity levels for the main naturally occurring radionuclides measured in suspended matter (top) and filtered waters (bottom) in the lower Rhône; 2002–2012.

Table 1. Detection frequency of artificial radionuclides in filtered water in 2012 (only radionuclides that were detected at least once are included).

Radionuclide	Detection frequency in 2010
^{137}Cs	100%
^{90}Sr	100%
^{3}H	100%
$^{239+240}$Pu	100%
^{241}Am	100%
^{60}Co	92%
^{238}Pu	69%
^{124}Sb	55%
^{125}Sb	25%
^{58}Co	25%
^{131}I	8%

dissolved phase (drinking water) and the particulate phase (suspended matter and sediments). In the River Rhône, the proportion of naturally occurring ^{14}C was between 50 and 70%. The proportion of naturally occurring tritium, however, was very small compared with artificial contributions (<5% on average) (Eyrolle-Boyer *et al.*, 2012).

2.2.1.2 *Artificial radionuclides*

Of the artificial radionuclides targeted in 2012, ^{137}Cs, ^{90}Sr, ^{3}H, $^{239+240}$Pu, ^{241}Am, ^{60}Co, ^{238}Pu, ^{124}Sb, ^{125}Sb, ^{58}Co and ^{131}I, listed in decreasing order of detection frequency, were detected in filtered water from the lower Rhône (Tab. 1). Systematic searches were not conducted for ^{3}H and ^{14}C. They can, however, be detected in the river by searching for them using certain analysis techniques. Searches were carried out for extremely low levels of transuranium elements ($^{239+240}$Pu, ^{238}Pu, ^{241}Am and ^{244}Cm) in environmental samples using alpha spectrometry measurements. They were almost always quantified (detected). For ^{3}H, activity levels varied from 5.6 ± 0.4 Bq/L to 10.4 ± 0.3 Bq/L, for ^{90}Sr

from 0.00062 ± 0.00002 Bq/L to 0.0019 ± 0.00002 Bq/L, and for ^{137}Cs, ^{124}Sb, ^{125}Sb, ^{58}Co and ^{131}I from 0.000019 ± 0.000002 Bq/L to 0.00052 ± 0.00001 Bq/L. Activity levels for transuranium elements ($^{239+240}$Pu, ^{238}Pu, ^{241}Am and ^{244}Cm) were between one and more than three orders of magnitude lower than the last-mentioned range (Fig. 3).

The detection frequencies and activity ranges for artificial radionuclides observed in 2010 were similar to those recorded in previous years.

Over the period 2002–2012, activity concentrations for all artificial radionuclides measured in filtered water from the lower Rhône tended to decrease by a factor of 5 to 10, except for tritium and ^{131}I, for which activity concentrations remained relatively stable.

Over the same period (2002–2012), HTO (tritiated water) activity levels were around 10 Bq/L (from 1.0 ± 0.7 Bq/L to 19.4 ± 1.1 Bq/L of combustion water, with an average of 5.6 ± 4.8 over the period 2010–2011), while activity levels for other artificial and naturally occurring radionuclides fell short of 1 mBq/L (Fig. 5). Activity levels for ^{14}C in the lower Rhône were estimated at 0.004 Bq/L (for an average carbon concentration of 20 mg/L), *i.e.* ten times higher than those for ^{90}Sr, an artificial beta-emitting radionuclide that is predominant in the lower Rhône (Eyrolle-Boyer *et al.*, 2012).

2.2.2 Activity levels measured in suspended matter

2.2.2.1 Naturally occurring radionuclides

As observed with filtered water, ^{40}K and the various elements in the ^{238}U and ^{232}Th decay chains, such as ^{234}Pa and ^{228}Ac, accounted for most of the natural radioactivity measured in the River Rhône (Fig. 3). Specific activity levels for ^{7}Be, a short-lived cosmogenic radionuclide with a half-life of 53 days, were significantly higher than those of other elements in the uranium/thorium decay chains. This element made a significant contribution to the natural radioactivity of particles transiting in the river. As in the case of filtered water, activity levels for ^{7}Be varied quite widely as they depend on the age of water and sediment "mass" in transit, as well as on precipitation over the drainage basin. ^{22}Na, which was found in filtered water, was not detected in suspended matter, because sodium is mostly found in dissolved form in continental water systems (Fig. 3).

As observed in filtered water, activity levels for naturally occurring radionuclides measured in suspended matter in the lower Rhône, in this case by gamma spectrometry, were stable over time and on the scale of observation concerned here, and were consequently the same as values recorded in previous years. Attention should, however, be drawn to seasonal variations in activity levels for some telluric elements, such as those observed here for ^{228}Ac. These are directly related to the types of minerals in transit, which depend on hydrology (Laubenstein and Magaldi, 2008; Eyrolle *et al.*, 2012).

2.2.2.2 Artificial radionuclides

Artificial radionuclides routinely sought and detected in 2012 in suspended matter in the lower Rhône

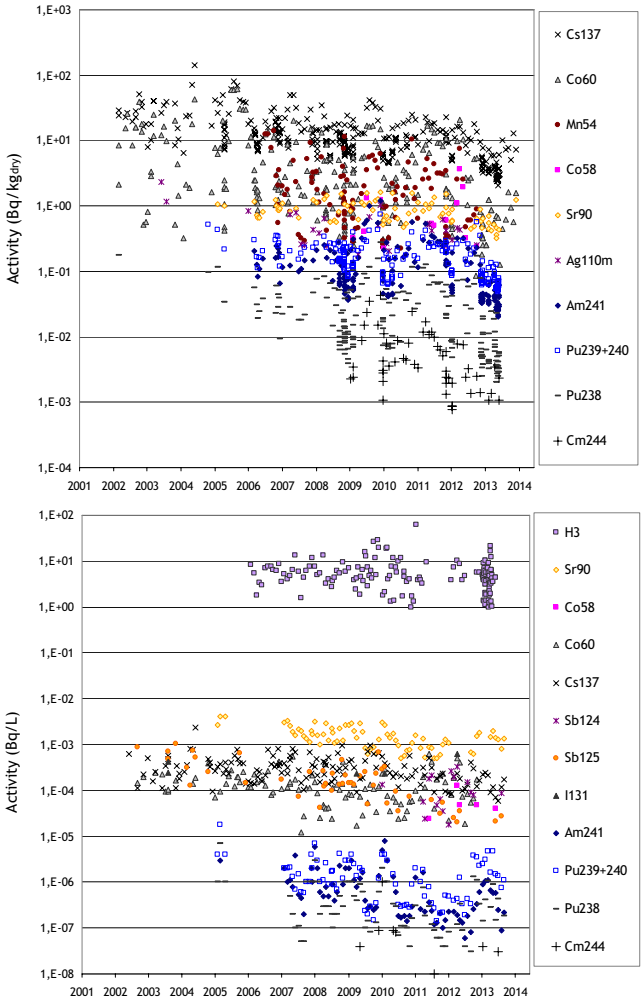

Fig. 4. Record of activity levels for the artificial radionuclides detected in suspended matter (top) and filtered waters (bottom) in the lower Rhône; 2002−2012.

were, in decreasing order of detection frequency, ^{137}Cs, $^{239+240}$Pu, ^{241}Am, ^{238}Pu, ^{60}Co, ^{90}Sr, ^{244}Cm, ^{54}Mn, ^{58}Co and ^{110m}Ag. As was the case with filtered water, transuranium elements ($^{239+240}$Pu, ^{238}Pu, ^{241}Am and ^{244}Cm) were measured by alpha spectrometry following radiochemical extraction and purification. They were detected at extremely low levels in suspended matter in water and very regularly quantified (Tab. 2). As in previous years, ^{137}Cs was the predominant artificial radionuclide measured in suspended matter (Fig. 4). Its specific activity varied from 3.2 ± 0.4 Bq/kg$_{dry}$ to 16.5 ± 1.6 Bq/kg$_{dry}$. Activity levels for other artificial radionuclides, measured by gamma spectrometry (^{60}Co, ^{54}Mn, ^{58}Co and ^{110m}Ag) or following beta radiochemistry (^{90}Sr) were up to two orders of magnitude lower. Note that activity levels for ^{14}C, a radionuclide of partly artificial origin, were higher than those of other artificial radionuclides and varied from 111 ± 2 to 294 ± 2 Bq/kg C (Fig. 5). Specific activity levels for transuranium elements ($^{239+240}$Pu, ^{238}Pu, ^{241}Am and ^{244}Cm) were between 0.001 Bq/kg$_{dry}$ and 0.1 Bq/kg$_{dry}$, *i.e.* lower than the above ranges of values. During the second half of 2012,

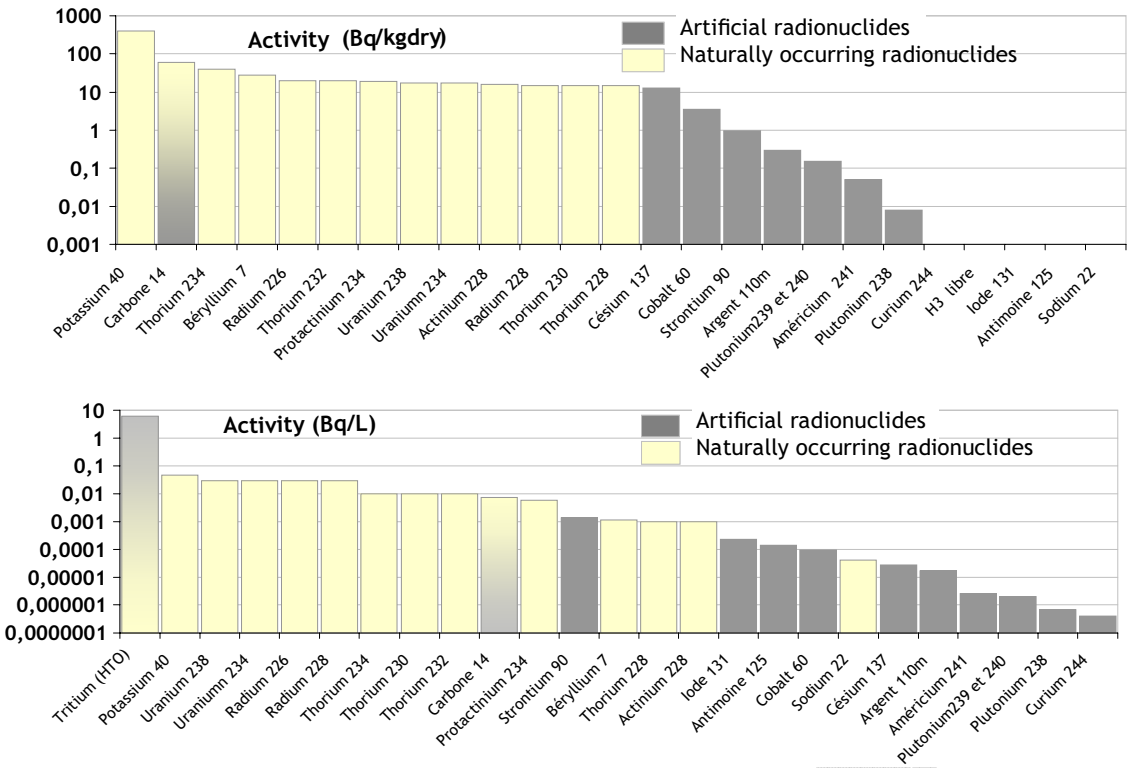

Fig. 5. Activity levels of the main naturally occurring and artificial radionuclides detected in suspended matter (top) and in filtered water (bottom) in the lower Rhône: SORA Station (2002−2012); tritium, iodine-131, antimony-125 and sodium-22 are elements that almost never bind to particles.

Table 2. Detection frequency of artificial radionuclides in suspended matter in 2012 (only radionuclides that were detected at least once are included).

Radionuclide	Detection frequency in 2012
^{137}Cs	100%
$^{239+240}$Pu	100%
^{238}Pu	100%
^{241}Am	100%
^{60}Co	97%
^{90}Sr	78%
^{244}Cm	38%
^{54}Mn	32%
^{58}Co	18%
^{110m}Ag	11%

activity ranges for some artificial radionuclides, especially ^{54}Mn and ^{60}Co, appeared significantly lower than those observed earlier in the year or in previous years. High-water levels in the Rhône generally led to a drop in the specific activity of most artificial radionuclides. This can be largely explained by the dilution of suspended matter contaminated by industrial discharge with telluric particles that are less contaminated, if at all (Eyrolle *et al.*, 2012). This trend cannot be explained solely by the floods in autumn 2012, as relatively low levels were also recorded outside this period.

Over the period 2002-2012, activity concentrations for all artificial radionuclides measured in suspended matter tended to decrease by a factor of 5 to 10 in most cases, as seen in filtered water (Fig. 4).

Most radioactivity in suspended matter in the lower Rhône was of natural origin. Levels for carbon-14, which is partly artificial in origin, were higher than the great majority of natural radionuclides (Fig. 5).

Lastly, it should be noted that the contribution of suspended matter to the overall radioactivity of river water, whether natural or anthropogenic, is of course dependent on the suspended load, particularly the river's rate of flow. In the lower Rhône, the suspended load increased by a factor of about 10 when the rate of flow increased from 2000 to 6000 m^3/s, thus modifying the proportions of radionuclides in transit in the dissolved and particulate phases (Antonelli, 2008, 2010, 2011, 2013).

3 Conclusions

Since the middle of the last century, nuclear facilities located in the Rhône valley have discharged low-level radioactive liquid effluent into the River Rhône.

The SORA monitoring station in Arles, located 45 km upstream of the mouth of the Grand Rhône, has continuously collected samples of large volumes of water to detect any trace concentrations of artificial radionuclides. The station

supplements the network that was set up along the Rhône corridor from the mid-1970s, to monitor the water in the River Rhône.

The resulting records show that the activity levels of artificial radionuclides in the Rhône, whether in the dissolved or particulate phase, have tended to decline, including over the last ten years. As expected, activity concentrations of naturally occurring radionuclides are stable over time (on a human scale).

Our research highlights the fact that most of the radioactivity in the River Rhône is of natural origin. However, tritium, of which a considerable proportion in the lower Rhône is due to nuclear industrial effluent, is the predominant radionuclide in filtered water. The same is true for ^{14}C regarding suspended matter.

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