The CAROL project (Camargue-Rhône-Languedoc)

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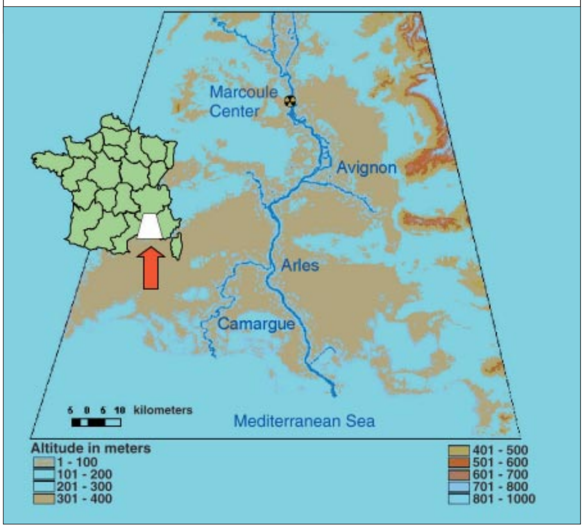
The initial aim of the CAROL project, launched in 1988, was to study and quantify the distribution of artificial radionuclides in one of the world's most nuclear-intensive environments: the lower Rhone Valley (**figure 1**). In the light of its findings, the growing concern of research workers, and current events, the study zone has been enlarged, primarily to take in Mercantour and Corsica.

Of the ten or so artificial radionuclides regularly present in measurable quantity in all the environment's components, the spotlight of the first four years' research work was focused on plutonium isotopes 234, 239 and 240, americium and cesium-137.

Figure 1

Location of the study catchment area.



1 - Because they are usually measured by alpha-spectrometry, the activity levels of ^{239}Pu and ^{240}Pu cannot be distinguished as their energy levels are very similar to those of alpha particles.

Actinides (plutonium-238, 239, 240 and americium-241)

In the soil

The research carried out on plutonium and americium has identified two origins for these actinides in the lower Rhone Valley environment (Renaud et al 2000, Duffa 2001):

- legacy fallout from the atmospheric nuclear arms tests carried out from 1945 to 1980 and the explosion of an American satellite with a payload of ^{238}Pu on board in 1964;
- discharges from the Marcoule nuclear center.

The distribution of these actinides in the soil is fairly even, with surface activity levels around 50 Bq/m^2 for ^{239}Pu + ^{240}Pu[1] and 1.5 Bq/m^2 for ^{238}Pu and ^{241}Am. Excluding an area of about thirty square kilometers around the Marcoule site where higher plutonium concentrations have been discovered, this can be attributed to legacy fallout. The total activity distributed over this area has been assessed at:

- 3 gigabecquerels (1 GBq = 10^9 Bq) of plutonium, representing a mean surface activity of about 120 Bq/m^2 instead of 50 Bq/m^2;
- 0.6 GBq of ^{241}Am, representing 24 Bq/m^2. The mean surface activity of ^{241}Am in France is about 20 Bq/m^2.

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Figure 2

Distribution of plutonium-239 and 240 in the soil around Marcoule.

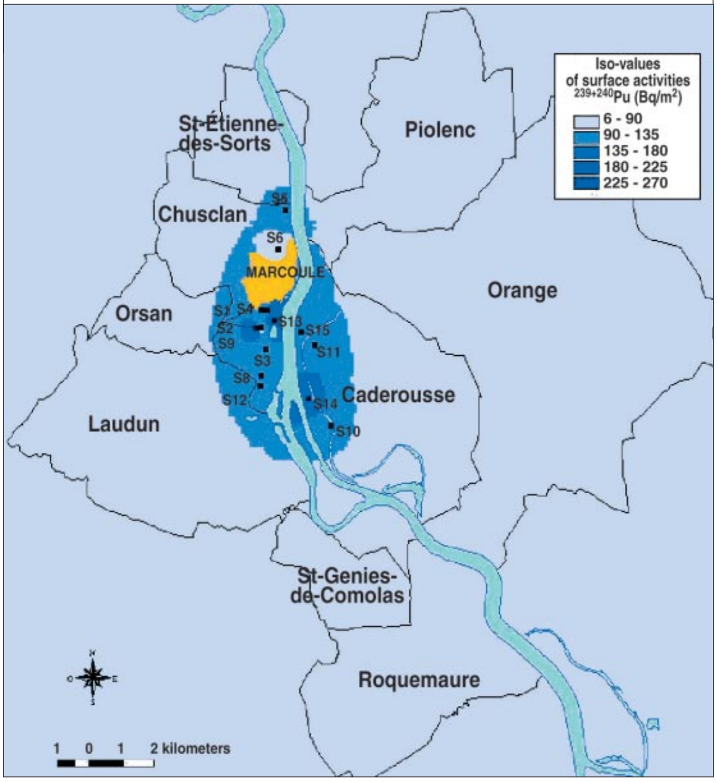
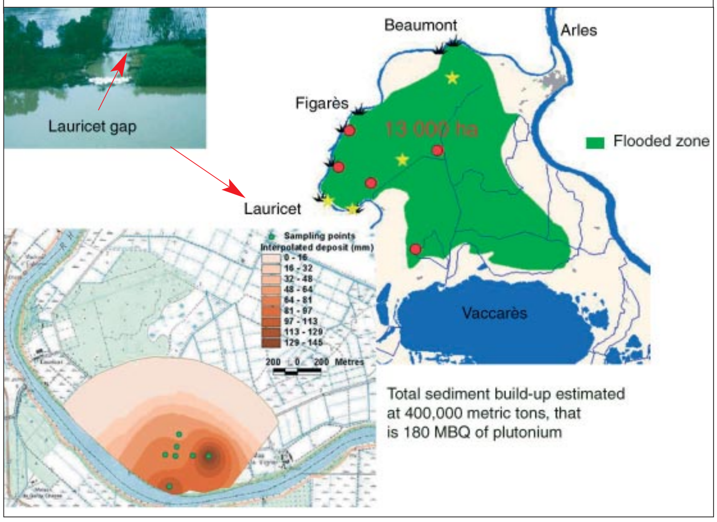


Figure 3

Location of flooded areas in 1993 and 1994 and dyke breaches, estimates of sediment depositions and the activity of the deposited plutonium.



This additional plutonium activity results mainly from atmospheric discharges pre-dating 1975, when weapon-grade plutonium was being extracted at Marcoule.

The map in **figure 2**, plotted from various measurements taken in the environment, details the distribution of plutonium-239 and 240. Similar maps have been plotted for plutonium-238 and americium-241.

The respective activity of the two potential origins of the actinides in the soil can be worked out from the plutonium-238 and plutonium-239 and 240 isotope activity ratio. Legacy fallout is characterized by a $^{238}Pu/^{239+240}Pu$ activity ratio of about 0.03. Soil measurements taken around Marcoule to take in the discharges due to the extraction of weapon-grade plutonium, bring this ratio up to 0.05. Moreover the same area witnessed more recent discharges up to 1998 from spent fuel reprocessing. They are characterized by a much higher activity ratio (plutonium at 0.3) that labeled specific plants such as mosses, thyme and grapes without significantly increasing their activity levels.

Now that reprocessing operations have come to a halt at Marcoule and the quantities of plutonium found in the soil are very low, they cannot lead to the local inhabitants being significantly exposed (the annual doses are calculated at under 0.001 mSv). The plutonium and americium readings for 1999 and 2000 in local crops taken with the best available measuring techniques, are at extremely low levels, in fact at the lowest detectable limits of activity.

Traces of fresher discharges originating from the Marcoule facilities are also discernible in soil irrigated by water from the River Rhone or high floodwaters. We have used the activity ratios calculated from readings taken close to the dyke breaches caused by the 1993 and 1994 Rhone floods to reconstruct the thickness of sediment deposited and extrapolate an estimate of the total volume of sediment deposited (**figure 3**).

In the Rhone

Discharges from Marcoule are seldom discernible in the terrestrial environment. This contrasts with the Rhone and its canals, which have been very regularly labeled with liquid discharges from this center. Until very recently, 90% of the plutonium conveyed in the Rhone river originated from the plant. Now that reprocessing has ended and discharges have been reduced, the main stock, and thus the main source of plutonium, is the build-up of sediment in the Rhone, its canals and its delta running into the sea. When water levels are high, sediment in the Rhone is a major source of plutonium in the water, second only to drainage of the catchment basin soils, and in front of the discharges from the Rhone Valley nuclear facilities (**figure 4**) (Eyrolle, 2001).

Figure 5 presents an inventory of the plutonium stocks and flows in the lower Rhone Valley over the past forty years. Most of this plutonium, that is 770 GBq, originates from nuclear arms test legacy fallout The atmospheric discharges from Marcoule only account for a small part (28 GBq), which is only visible in the immediate vicinity of the site. However the plant accounts for 90% of the plutonium (920 GBq) that has moved along the Rhone River over the past forty years. The additional activity in the soil resulting from irrigation or high floodwaters is at very low levels and only detected near dyke breaches during flooding.

Cesium

Reconstructing cesium-137 depositions from 1986

The cesium-137 in the lower Rhone Valley environment, as for the whole of Eastern France, primarily originates from fallout from the Chernobyl accident, which was very patchy as rainfall was instrumental in its deposition. Deposition increased with rainfall over the first week in May 1986 (**figure 6**) (Renaud et al, 2001). Certain sites in the Vaucluse were singled out for research into this cause-effect relationship because they had heavy rainfall in May 1986 and secondly because their clay-rich soil has retained most of the cesium. Cesium-137 deposition for the whole of Eastern France has been mapped (**figure 7**, page 204) on the basis of this rain/deposit relationship and the precipitation depths recorded by Météo France in May 1986.

This map highlights how uneven radioactive deposition is and shows that the regions that had precipitation depths of over 20 mm record the highest depositions.

It was estimated in 1997 that 2300 communes were affected by higher-than-average deposition levels for their departments, in the range 3000 - 6000 Bq/m^2 for the farming plains of Eastern France. The deposition levels of 180 of these communes were in the range 25,000-35,000 Bq/m^2.

Figure 4

Origin of the plutonium found in the Rhone River in line with waterflow.

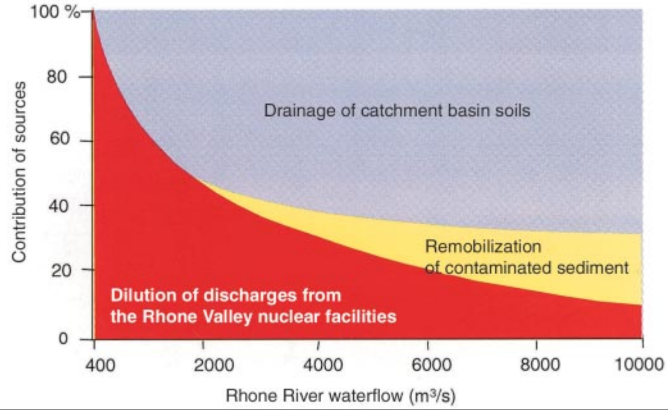


Figure 5

Plutonium stocks and flows in GBq in the lower Rhone Valley over the past forty years.

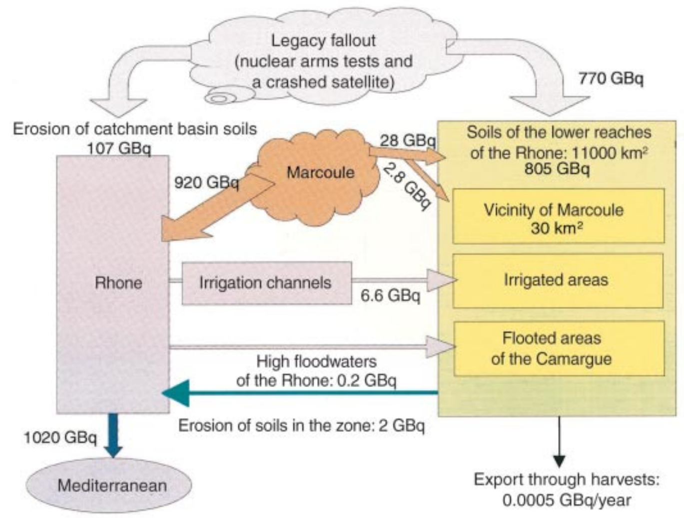


Figure 6

Relation between the cesium-137 deposition levels and rainfall during the first week of May in 1986.

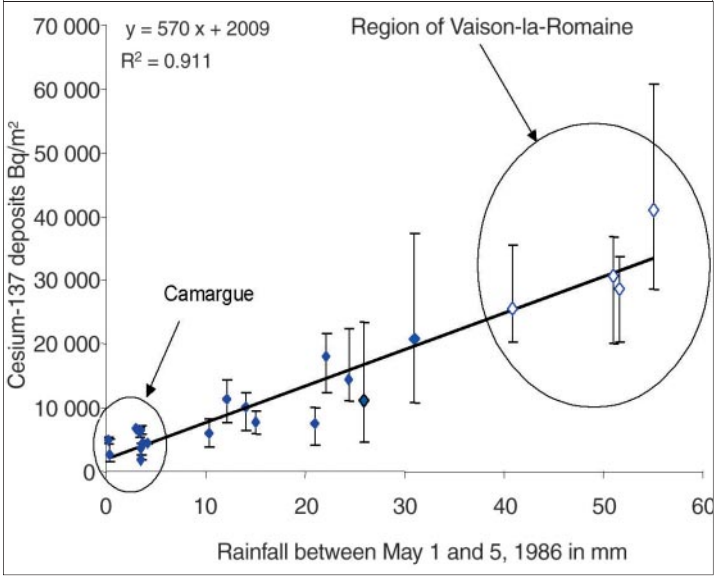
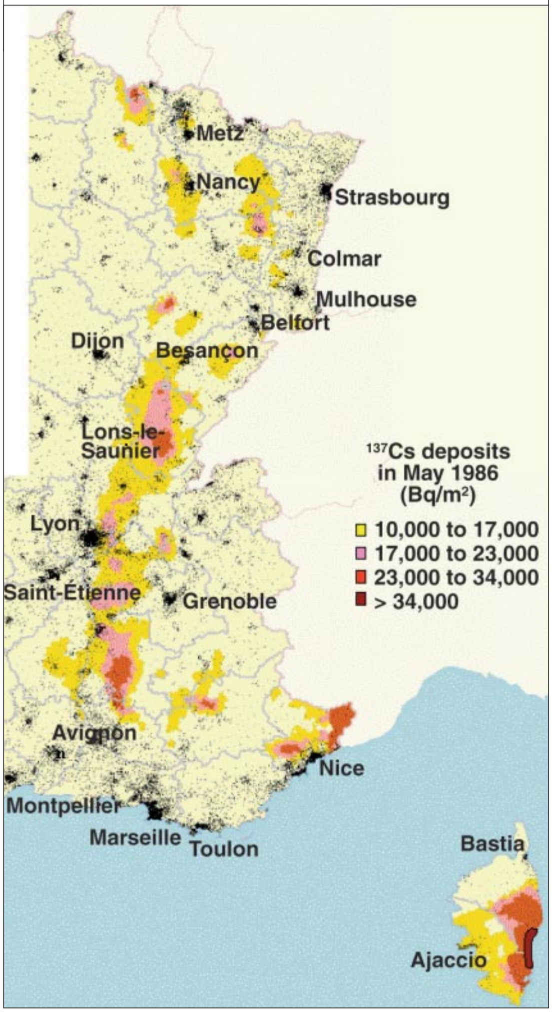


Figure 7

Distribution map of cesium-137 deposition in May 1986.



Iodine-131 depositions were at levels five to ten times higher than those of cesium-137 (Renaud & Métivier, 2000).

TODAY'S CESIUM-137 DEPOSITIONS

• Mercantour

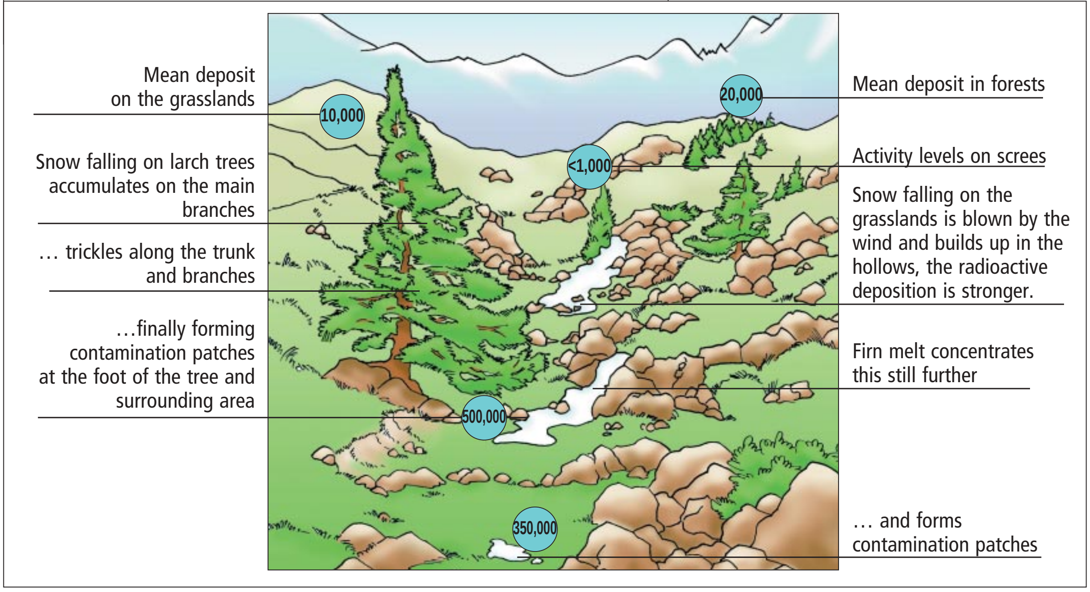
A specific study has been made to explain the "hot spots", namely the cesium-137 patches with concentrations in excess of 100,000 Bq/m^2 over several square decimeters to several square meters in the mountains (Pourcelot et al, 2001). They turned out to be deposit accumulation points in the snow dating back to May 1986 and reconcentration at the end of the spring when the firn melted. Other atmospheric pollutants are also concentrated at these points: lead from fuel, antimony from vehicle braking systems, copper, nickel, cadmium…(**figure 8**).

• Comtat Venaissin (part of the Vaucluse)

Certain clay-rich soils such as those in Comtat Venaissin or Haute Provence have preserved high levels of the cesium-137 deposited in May 1986 to this day, whereas in Corsica only residual amounts remain. A soil study has shown that in Comtat Venaissin, over 80% of the cesium-137 is trapped in the first 10 centimeters of topsoil clay and that migration rates are very low. Although this region was deeply affected by fallout from the Chernobyl accident, the cesium-137 activity in

Figure 8

Location and origin of the contamination "patches" in the mountains.



wine has now reverted to its pre-Chernobyl level. In fact **figure 9** shows that the 1986 vintage was slightly affected by the radioactive depositions because at the beginning of May the vines were at an advanced stage of development. As the cesium-137 taken up by the woodstocks leached out, the activity level of the wine dropped rapidly. The activity level recorded in 2000 is similar to that of 1983. This is explained by the fact that most of the cesium deposited in the wake of the Chernobyl accident remained on the soil's surface. It is still not available for uptake by the vine roots and will not be ready for over fifty years. By then 80% of the activity initially deposited will have been depleted by radioactive decay (Renaud et al, 2001).

• Corsica

Preliminary results of the three soil sampling campaigns carried out in Corsica in 2001 by IRSN confirm that the island's east coast and mountain massifs are some of the areas of France most affected by fallout from the Chernobyl accident. However the residual cesium-137 activity in the area is very patchy due to the rainfall in May 1986, and most of all to the pedological characteristics of the soil that have retained in various ways these depositions. In certain cases, over half the cesium-137 has disappeared through soil drainage and flown into the sea via the groundwater and streams (Pourcelot et al, 2002).

• The submerged delta of the Rhone

Soil drainage in the submerged delta of the Rhone (**figure 10**) has led to a build-up of 6 TBq of cesium-137 trapped in the sediment over a relatively small area of 160 km^2. This concentration is compounded primarily by 10 TBq from the Rhone Valley nuclear facility discharges, which accounts for 20% of the radioactivity released into the Rhone since 1961 (Charmasson, 1998). The largest accumulation of cesium-137 in the lower Rhone Valley, a total of 20 TBq, lies in the submerged Rhone delta. With values in places exceeding 400,000 Bq/m^2, the sporadic activity concentrations present are much in line with the contamination patches of Mercantour, rather than the concentrations found in the valley soils that are never above a tenth of this value. The explanation for this is that over 80% of the cesium in the aquatic environment is taken up by particles in suspension (less than 20% in solutes), these being particles that have settled in the

Figure 9

Activity concentration of Côtes du Rhône wine.

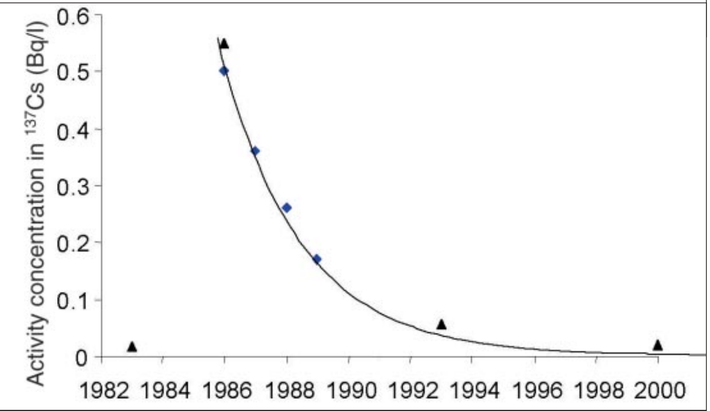
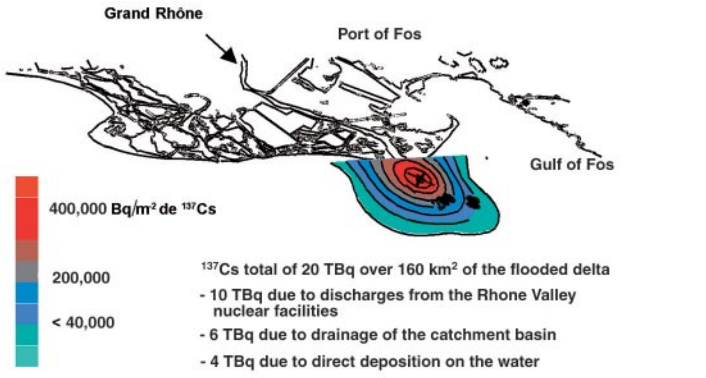


Figure 10

Distribution, stock and origin of cesium-137 in the submerged Rhone delta.



Corsica's east coast and mountain massifs are some of the areas of France most affected by fallout from the Chernobyl accident

submerged delta. Following on from cesium, research is currently being conducted into the accumulation of actinides (plutonium and americium) and the future of these radionuclides trapped in this way.

Prospects

At the end of its fourth year, the CAROL project has answered the key questions about the origin of the radioactive nuclides present in the lower Rhone Valley environment, and has provided an inventory of the current stocks and modeling of the uptake and exchange flows of its various components for the five radionuclides covered by the research: ^{137}Cs, ^{238}Pu, ^{239}Pu, ^{240}Pu and ^{241}Am. Apart from completing the work in progress, by assessing the actinide stock in the prodelta, this program could be broadened to include other radionuclides, particularly ^{129}I and ^{90}Sr and study of anthropogenic flows, which have not been addressed up to now. A more detailed study of certain mechanisms should extend the first part of the project, which enabled general effects to be quantified: chemical splitting during radionuclide migration through soil, competitions between stable and radioactive elements, radionuclide speciation in freshwater, and the importance of the resuspension of soil particles in transfers to vegetation.

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