Impact of uranium mines water treatment on the uranium and radium behaviour

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Abstract Uranium has been mined in the Limousin area (France) until the end of the 90's. Nowadays mines are closed and environmental monitoring is conducted by the former operator (AREVA-NC) under the control of the French administration. In the year 2006, the French safety authority board has created a pluralist expert group (GEP) that enjoin experts, stake holders and government representatives, to improve the environment protection concerning the process of uranium mines closure and rehabilitation. A part of the work is devoted to the study of mines as radionuclides sources and to the transport of radionuclides in the environment. According to regulation, site waters must be collected, and when necessary treated, before being released into the environment. Those waters constitute the main vector of radionuclides transportation. Several previous studies revealed an accumulation of radionuclides in lake sediments downstream mines inputs. This phenomenon is illustrated on the Ritord watershed; in an artificial lake created only few kilometres downstream one of the main treated mine waters release. To identify the mechanisms of the radionuclides accumulation in sediments, the GEP has investigated, through the environmental survey database, the water treatment efficiency and the behaviour of ^{238}U and ^{226}Ra from mines to the river. It concluded that a part of particles containing ^{226}Ra produced in the water treatment station is not trapped but released to the river. The reduction of the water flow velocity where the stream enters the lake leads to the particles

deposition. As a consequence, the water treatment station has been modified to reduce the particle outflow. The uranium and radium content on particulate, colloidal and dissolve fractions were then investigated over a year all along the treatment process as well as downstream the water release point. Results of this specific study will be presented.

Introduction

Uranium has been mined in the Limousin area (France) until the end of the 90's. Nowadays mines are closed and restored. Rehabilitation operations were devoted to assure the stability of land and infrastructures as well as to the collection of mine's waters to keep under control the radionuclide release into watersheds. Environmental monitoring is conducted by the former operator (AREVA-NC) under the control of the French administration which defines the objectives of environmental and population protection against radiations. Concerning natural radionuclides from former uranium mining installations, the French Decree 90/222 indicates that dissolved ^{226}Ra and ^{238}U activities in water released into the environment must remain under the limits of 0.74 Bq.l^{-1} and 22.4 Bq.l^{-1}, respectively. The waters flowing out the mines are collected and analysed to determine their ^{226}Ra and ^{238}U contents. When necessary, waters are treated before being released.

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This phenomenon is illustrated on the Ritord watershed in an artificial lake created only few kilometres downstream mines. To identify the mechanisms of the radionuclides accumulation in sediments, the GEP has investigated, through the environmental survey database, the water treatment efficiency and recommended to the operator to conduct a complementary study to determine the speciation of ^{238}U and ^{226}Ra from mines to the lake. The work was conducted between October 2006 and October 2007. Results are presented conjointly with previous investigations hereafter and interpreted from the management options point of view.

Area

About 73 000 tons of uranium were extracted from the 200 French uranium mine sites. The Crouzille mining division (~ 300 km²) produced 24 000 tons of uranium from 24 underground and open mines. Seven watersheds compose the hydrological web of the Crouzille Division area. Among them, the Ritord basin (figure 1) was selected to illustrate the impact of mine's waters outflow. It corresponds to a small brook connecting two lakes. The upper one named "Gouillet" could be considered as not influenced by mines. Its outflow is the Ritord stream witch collects water from seven mine sites. Waters from St Sylvestre, Fanay-Augères and Silord are treated before being released into the environment due to their ^{226}Ra activity level higher than the authorized limit of 0.74 Bq.l^{-1}. Waters from Borderie and Vénachat do not require any treatment and no water flowing out the Bachellerie and Santrop sites was identified. It can be seen on figure 1 that in term of uranium production, the most important production comes from the Fanay-Augères site. The Ritord flows into the "Saint Pardoux" lake created in 1976 by the building of a dam on the river Couze flow about 1.5 km downstream the Couze/Ritord confluence.

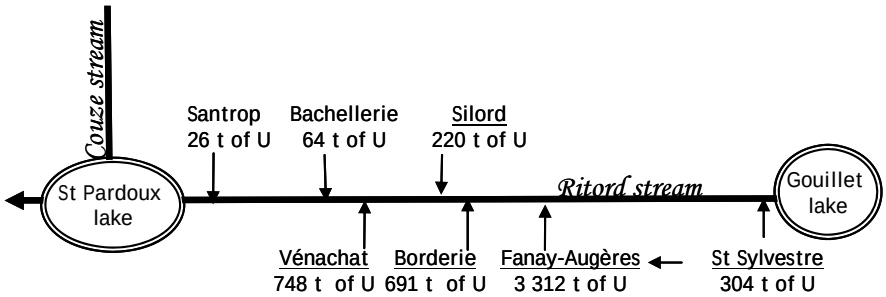

Fig.1. Production of uranium from mines in the Ritord watershed

Impact of mine's waters inflow to the Ritord

On the base of the survey operated by AREVA-NC under the control of the French administration (waters) or on itself decision (sediments), the impact of the mine's waters outflow in the Ritord brook was investigated in terms of radiological content.

Figure 2 presents, on its upper part, the dissolved ^{238}U (squares) and dissolved ^{226}Ra (lines) activities in waters from the Gouillet lake and all along the Ritord flow. On its lower part, ^{238}U, ^{226}Ra and ^{210}Pb activity levels in sediments are presented. The influence of each mine water inflow may be deduced from the

comparison of measurements conducted up and downstream the releasing point represented by dashed lines on figure 2. Waters were analysed monthly between 1990 and 2006; all data are reported except for the ones under the detection limit. The ^{238}U, ^{226}Ra and ^{210}Pb activity levels in sediments reported on figure 2 correspond to the mean values of measurements conducted near annually between 1992 and 2004.

Results on waters are analysed on the spatial and temporal dimensions. On the Gouillet lake, dissolved ^{238}U and dissolved ^{226}Ra activities fluctuate respectively around 0.01 $Bq.l^{-1}$ and 0.04 $Bq.l^{-1}$ with a more important variability for the ^{226}Ra. Downstream the St Sylvestre site, activity level increase slightly compared to the Gouillet lake. Downstream the Fanay-augères site activity levels reach the higher values measured on the Ritord stream. Then the Fanay-Augères site could be considered as the main source of radionuclides to the Ritord. This is confirmed by a mass balance along the Ritord flow which indicates that more than 90% of the ^{238}U and the ^{226}Ra entering the Saint Pardoux lake through the Ritord comes from the Fanay-Augères outflow. Influence of Silord, Borderie and Vénachat is limited compared to the Fanay-Augères one's. It can also be noted that the increase is more important for the ^{238}U than for the ^{226}Ra. Considering the temporal variation higher activities correspond to the beginning of the 90's. They are contemporary with the restoration works on the Fanay-Augères site. Nowadays, dissolved ^{238}U and ^{226}Ra activity levels in the Ritord's water -downstream the Fanay-Augères water release point- are both around 0.1 $Bq.l^{-1}$ and the main influence remained linked to the uranium release.

Results on sediments show that activity level of the three radionuclides analysed is higher on lakes than on the Ritord. This could be linked to the size of sediment's grains which is bigger on rivers than on lakes. As the specific surface is inversely proportioned to the size of grain, specific activity increase when the particle size decrease. As a consequence, activity levels in sediments are higher in lakes than in rivers. Considering the two lakes, the comparison of activity levels in sediments shows an important increase up to down stream. Sediments from Saint Pardoux lake were sampled near the Ritord entrance into the lake and higher values were fund in the literature (up to 20 000 Bq.kg-1 of 238U were measured). The increase of the activity level in sediment may be linked to the input by mine waters.

The increase of activity level in water and sediments may be considered as a potential radiological hazard. Apart from this aspect, the French authority asked to AREVA-NC to work on treatments to keep ^{238}U activity level in sediments of Saint Pardoux lake under the value of 3 700 $Bq.kg^{-1}$.

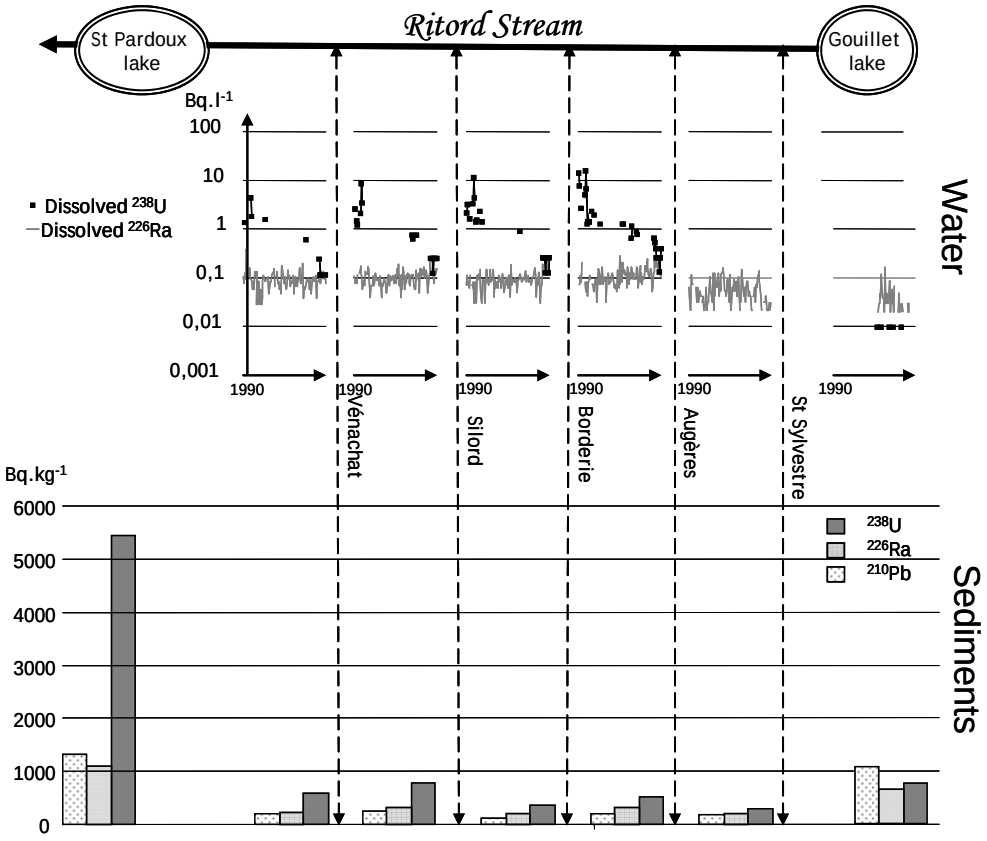

Fig.2. Influence of mine waters input on water and sediments radiological content in the Ritord stream and Saint Pardoux lake**.**

Efficiency of Mine's water treatment

As more than 90% of uranium and radium come from the Fanay-Augères treatment station, the study focus on it. The actual regulation impose to AREVA-NC to keep dissolved ^{238}U and dissolved ^{226}Ra activity in water flowing out the station respectively under 22.4 $Bq.l^{-1}$ and 0.37 $Bq.l^{-1}$. Then chemical treatment applied by AREVA-NC focus on dissolved ^{226}Ra which is transformed as insoluble specie by $BaSO_4$ precipitation before to settle in retention basins.

Figure 3 shows the Fanay-Augères mine's water treatment efficiency between 1993 and 2006. The first graph presents the activity level of dissolved ^{226}Ra in water entering and exiting the water treatment station. The difference between the

two corresponds to the efficiency of the transfer of dissolved ^{226}Ra on the particular form. The main yield over the duration of the study reaches 90%. Then the treatment seems very efficient. Nevertheless, the comparison between total ^{226}Ra entering and exiting the station shows that the total yield is 70%. That means a part of particles produced in the station does not settle in the basin and escape. They are released into the Ritord and may settle when they reach the lake. The last graph on the figure 3 presents activity levels of dissolved ^{238}U entering and exiting the treatment station. It shows that the chemical treatment applied is not efficient for uranium. In fact, as the dissolved ^{238}U activity level in water entering the station is under the legal limit of 22.4 Bq.l^{-1} (annual mean), no treatment is deployed for uranium.

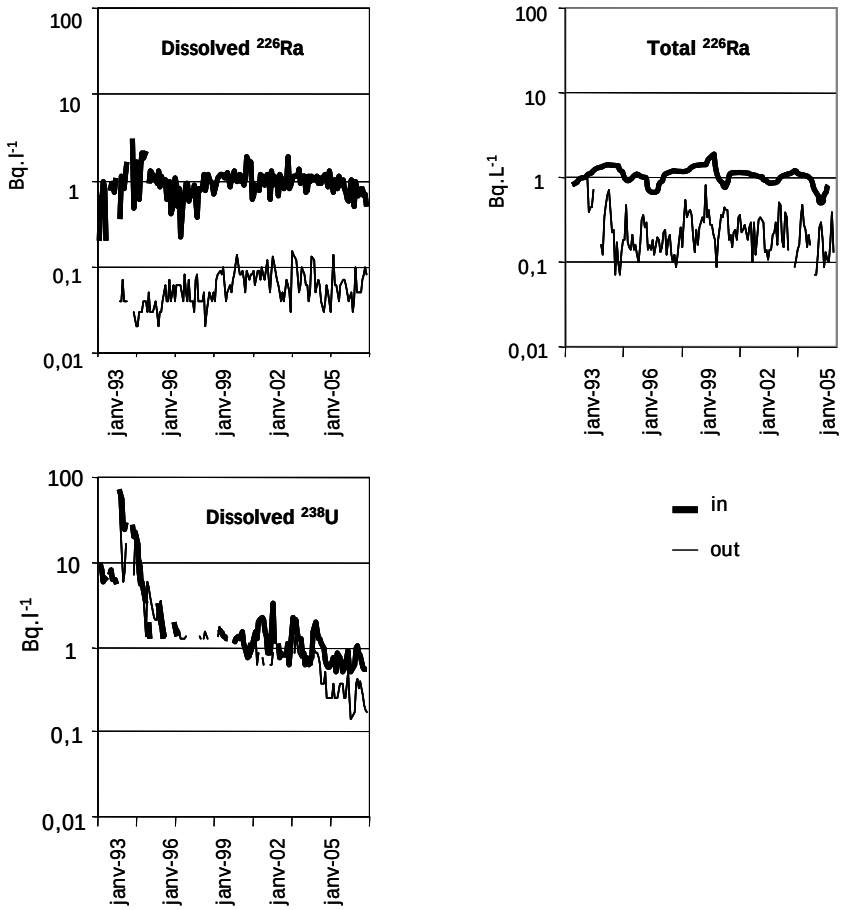

Fig.3. Fanay-Augères mine's water treatment efficiency

Nevertheless, as the objective is to reduce the ^{238}U activity level in sediments, developments were conducted to increase the settling in the basin and to remove uranium from water.

Efficiency of developments in the Fanay-Augères water treatment station

Developments proposed by AREVA-NC consist in building a new settling basin, using new chemical agents, and optimising the flow in the station to increase the residence time in the station. Waters flowing out the mines are collected in a basin (not represented on figure 4) and convoyed to the treatment line where chemical agents are added in the flow. Then waters pass through four basins before being released in to the Ritord flow. Basins 1 and 2 work in parallel on waters exiting the chemical treatment line. Waters exiting theses basins are directed to basins 3 and 4 in series. The efficiency of developments was assessed through the measurement of ^{226}Ra and ^{238}U in dissolved and particulate fraction in waters entering the station, all along the treatment (basin 1+2; basin 3 and basin 4) and downstream the release point in the Ritord. Results are presented in figure 4 with ones of a similar previous study conducted in 2001. At this time waters flowing out the chemical treatment line flowed through three basins in series. Waters from mines, from the last basin and from the Ritord downstream the release point were analysed in 2001. Results are presented on figure 4.

Results show a strong decreasing of uranium in water from mines over time. This trend is weaker for the ^{226}Ra. Activities of ^{226}Ra measured on the basin 4 during the year 2007 are lower than ones obtained on basin 3 during the year 2001. That attests the improvement of the water treatment yield on the ^{226}Ra. Indeed, in water from mines ^{226}Ra is on the soluble form. As mentioned above, the chemical treatment produces particles able to catch the radium. It appears that changes on the flow in the basins are efficient to increase the settling of particles. Most of them are retained in basins 1 and 2, but some settled in basins 3 and 4. On the opposite, a small proportion of uranium is trapped during the treatment and most of uranium remained on a soluble form when it reaches the Ritord flow.

Between 2001 and 2007, the concentration in uranium is divided by 3.2 in exit of treatment. It reverberates by a content of uranium divided by 2.4 in the Ritord flow. For radium, the concentration also decrease and are divided by 2 in water at the exit of basin of treatment and by 1.2 in the Ritord. The water samples of Ritord downstream Augères effluents have intermediate characteristics between two poles: The diluting pole: water of refill (rainwater having more or less interacted with the granitic substratum, and the typical pole of water of mine which is characterized by contents relatively rich in Ca, Mg, U, SO_4 and Ba (Ba is an agent

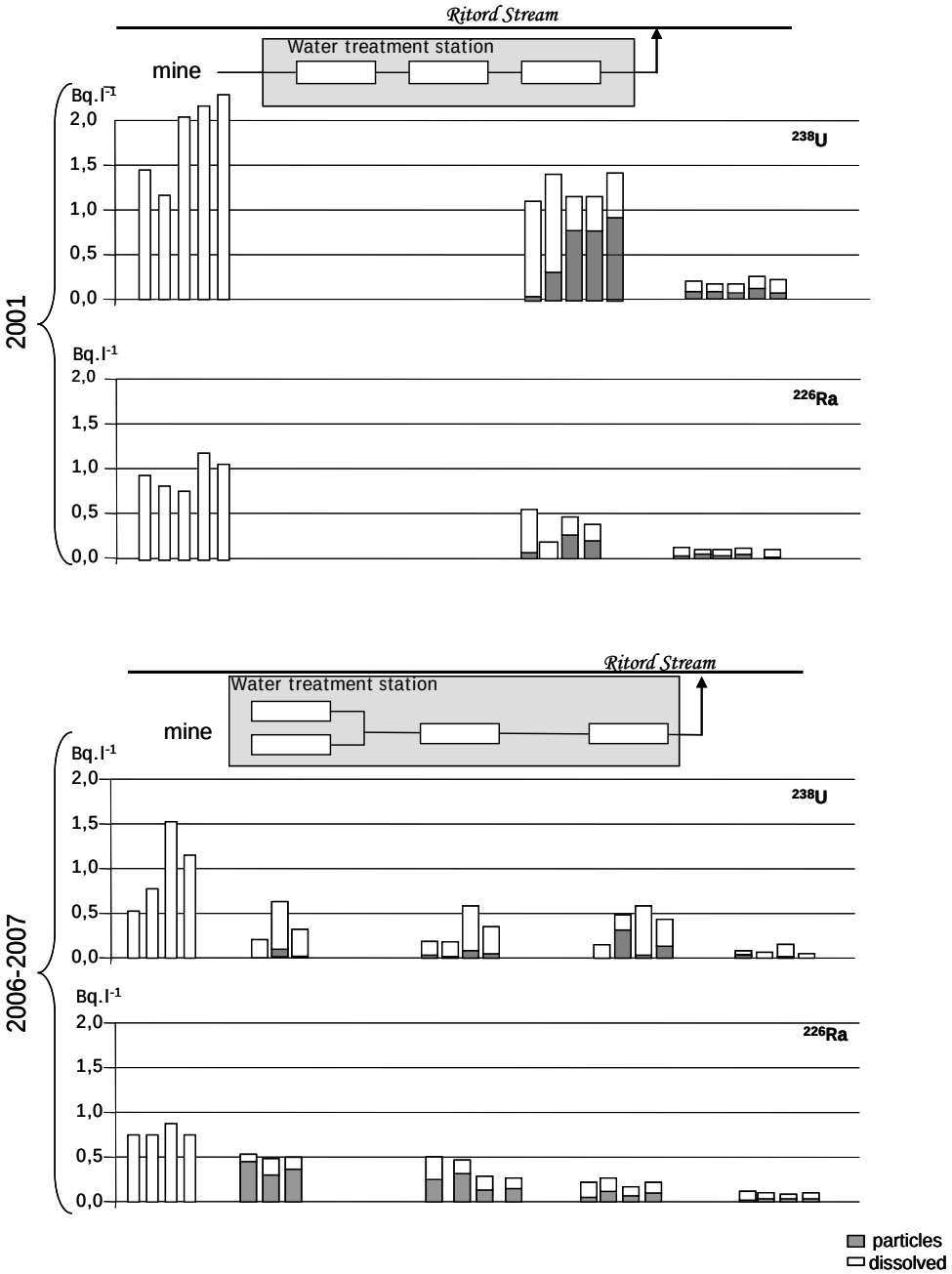

Fig.4. Distribution of ^{238}U and ^{226}Ra in water from mines and in the different water treatment steps before and after changes in the water treatment station.

of treatment of the mining effluents). The proportion of the pole "mining water" in waters of Ritord remains weak and decreases in a significant way since 2001.

Conclusion

Dissolved ^{238}U and dissolved ^{226}Ra activities in natural waters of the Limousin region (France), vary respectively around 0.01 $Bq.l^{-1}$ and 0.04 $Bq.l^{-1}$. Corresponding activities in waters flowing out former mines are around 1 $Bq.l^{-1}$. As these waters may, locally, constitute the main contribution of the flow, the radiological impact on the environment is monitored. In order to satisfy the French regulation chemical treatments are applied by the operator to limit the release of radionuclides.

That work indicates that activity levels in the environment depend on the flux released but also on the environment collecting and convoying flux. It shows the importance of lakes which presents the capability to trap radionuclides within their sediments. To limit the accumulation of “radioactives particles” in lakes, the functioning of the water treatment station was optimized. From the general point of view, the quality of the natural water of the Ritord increase. It results not only from changes operated in the station but also from the decrease of ^{238}U content in water flowing out the mines. Results of the monitoring of the Saint Pardoux Lake do not show a significant improvement in the radiological characteristics of the sediments, what demonstrates the efficiency limit of the chemical treatment.