Evolution of the natural radioactivity within the Soultz geothermal installation

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ABSTRACT

After the first studies on natural radioactivity done during the 2005 hydraulic circulation test at the Soultz-sous-Forêts power plant, the ASN (French National Agency for Nuclear Safety) recommended to follow precisely the evolution of natural radioactivity within the geothermal installation. The first goal of these studies is to ensure the protection of workers against potential radiations**.**

Seven measurement campaigns have been carried out since 2009 to observe and characterize the natural radioactivity evolution during hydraulic circulation tests, both on GPK2 and GPK1 surface installation. For all measurements campaigns, the results show a general increase of the dose rates with the circulation time and volume. The highest values were found mostly on the reinjection line, where the temperature is lower (~70°C). This indicates a correlation between the observed radioactivity and the scaling processes inside the installation: some newly formed minerals are able to trap radionuclides.

Finally, all these studies allowed us to take several measures for workers and public radioprotection. A restricted zone has been defined around the installation and adapted equipments must be worn by workers, when they have to operate on surface facilities, especially when dismantling pipes or cleaning heat exchangers and filters. Moreover, a proper plan has been established for the disposal of radioactive waste, consisting mainly of residues from filters: they have to be removed from the site, following the French regulation.

1. INTRODUCTION

Natural radioactivity becomes a growing concern among the geothermal community, especially on geothermal plants that exploits reservoirs hosted by rocks naturally containing radionuclides. This is, for instance, the case of geothermal power plants located in the Upper Rhine Graben, whose boreholes are often drilled down to the crystalline basement. Indeed granitic rocks contain radionuclides, mainly, Uranium, Thorium and products from their disintegration chain. Geothermal fluid, which naturally circulates in this fractured granite, is able to leach some radionuclides and bring them to the surface through pumping.

In 2005 a 6-months circulation test was performed at the Soultz site in artesian conditions. During this test, first measurements on the surface installation revealed the occurrence of natural radioactivity. Results were sent to the ASN (French National Agency for Nuclear Safety), which recommended to start a regular monitoring of the evolution of natural radioactivity on the surface installations. Despite the fact that the radioactivity level is rather low, the French nuclear regulation requires to set up radioprotection measures, as soon as radiations may occur, in order to ensure that any worker will not receive a cumulative dose larger than 1 mSv over a period of 12 consecutive months.

Since 2009, several circulation tests of different durations have been performed while testing the Soultz-sous-Forêts power plant. During each of them, at least one measurement survey has been made so as to observe the evolution of natural radioactivity on the surface installation and try to correlate it with hydraulic parameters. In parallel a research work has been launched in order to better understand the origin of radioactivity occurring in the surface installation. This paper presents the results of the surveys and of the scientific research, as well as the applied radioprotection procedures.

2. RADIOACTIVITY MEASUREMENTS

Between June 2009 and April 2012, 9 radioactivity measurement surveys have been performed on the Soultz geothermal installation during or after the circulation tests described in 2.2. They allowed monitoring rather precisely the evolution of the radioactivity level during a given test and to compare it from one test to the others.

2.1 Measurement method

A radiameter is used for the measurements (Fig. 1): it allows to record two types of parameters:

- **the activity**, which is the number of nuclear disintegration per second,
- **the dose rate**, which is the dose of radiation received by a body per unit of time. Here it is expressed in µSv/h (micro-Sievert per hour).

Figure 1: Dose rate measurement performed with the radiameter.

As the dose is the main relevant parameter used in radioprotection, dose rate measurements are performed. “Contact” data are recorded, that are taken 1 cm away from the installation. They are used to precisely map the radioactivity level on the different parts of the surface installation. “Ambient” values are also measured 1 m away from the installation: they are more representative of actual work places and used to set up radioprotection procedures. Around 350 contact and 50 ambient measurements are sampled both on GPK2 and GPK1 platforms. As they are precisely identified, measurements can be repeated at the exact same position, so as to compare values from one survey to the next.

GPK2 platform comprises the main geothermal installation (Fig. 2), with 3 5-km deep boreholes, GPK2 (production with a downhole pump), GPK3 (reinjection) and GPK4 (production, then reinjection). The 1.5 MWe power plant and all associated equipments (pipelines, heat exchangers, filters) are also located on GPK2 platform. On GPK1 platform are located only GPK1 borehole (reinjection) and the associated equipment (mainly pipes).

Figure 2: View of GPK2 platform

2.2 Circulation tests and corresponding surveys

9 radioactivity measurements surveys have been performed over 5 circulations tests:

- *circulation March-October 2009*: 2 surveys performed in June 2009, after 2 months of circulation and October 2009, after the end of the test
- *circulation November 2009 – October 2010*: 3 surveys performed in June 2010, after 6 months of circulation, in August 2010, after 9 months of circulation and October 2010, just before the end of the test
- *circulation January 2011 – April 2011*: 1 survey performed in April 2011, after the end of circulation
- *circulation August 2011 – October 2011*: 1 survey performed in October 2011, after the end of the test
- *circulation March 2012 – April 2012*: 1 survey performed in April 2012 at the end of the circulation test.
- *circulation January 2013 - ... (ongoing):* 1 survey performed in April 2013, during the circulation test (results not reported in this paper)

It has to be noted that another survey has been performed in July 2011, between both 2011 circulation tests.

2.3 Results from GPK2 platform

Figure 3 presents the evolution of dose rates at the 350 contact measurements positions for the 9 surveys described above. Figure 4 shows the same results, but for ambient measurements. For the latter, several measurements points have been added in April 2012, that were not checked during the former surveys.

In June 2009, the dose rates values (contact measurements) varied from the background noise level (~0.06 µSv/h) to a maximum of 4.90 µSv/h, with an average of 0.89 µSv/h (Cuenot, 2009; Maquet, 2011).

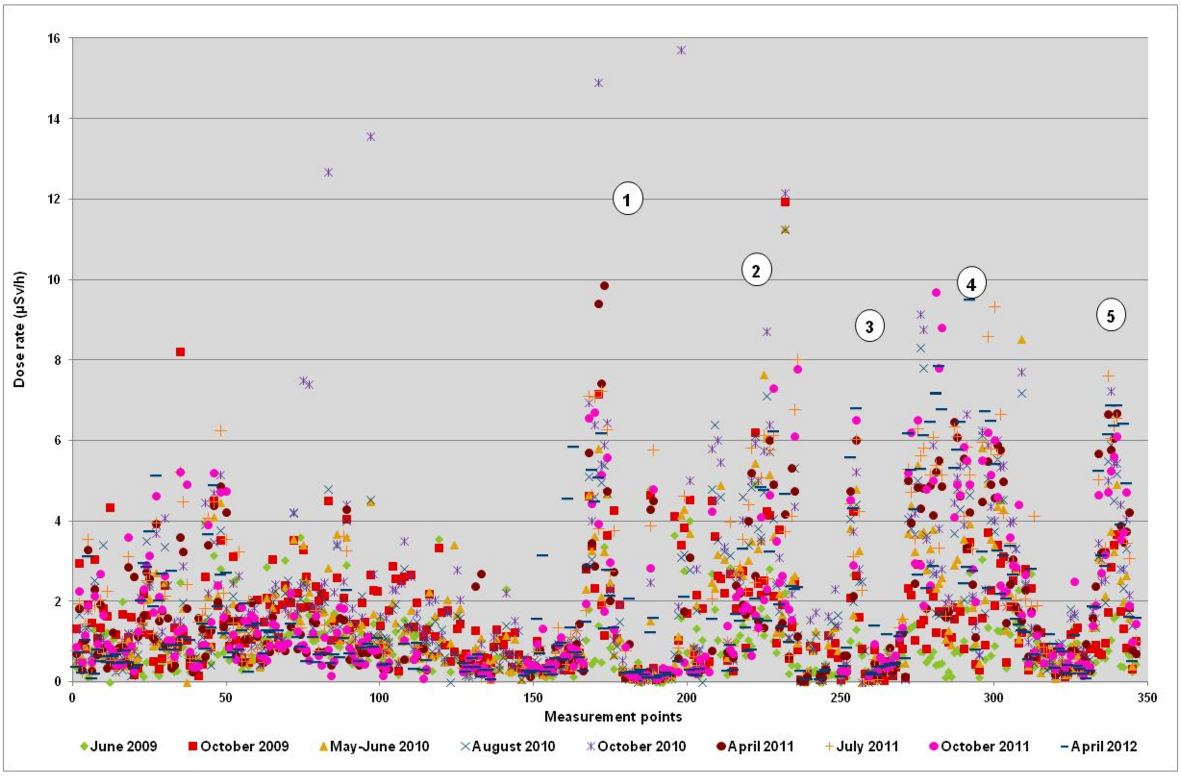

Figure 3: Evolution of contact dose rate measurements on GPK2 platform during the 9 surveys. The circled numbers correspond to: 1) Filters on reinjection lines, 2) Part of reinjection line, 3) reinjection line to GPK3, 4) Part of reinjection line and 5) Reinjection pumps

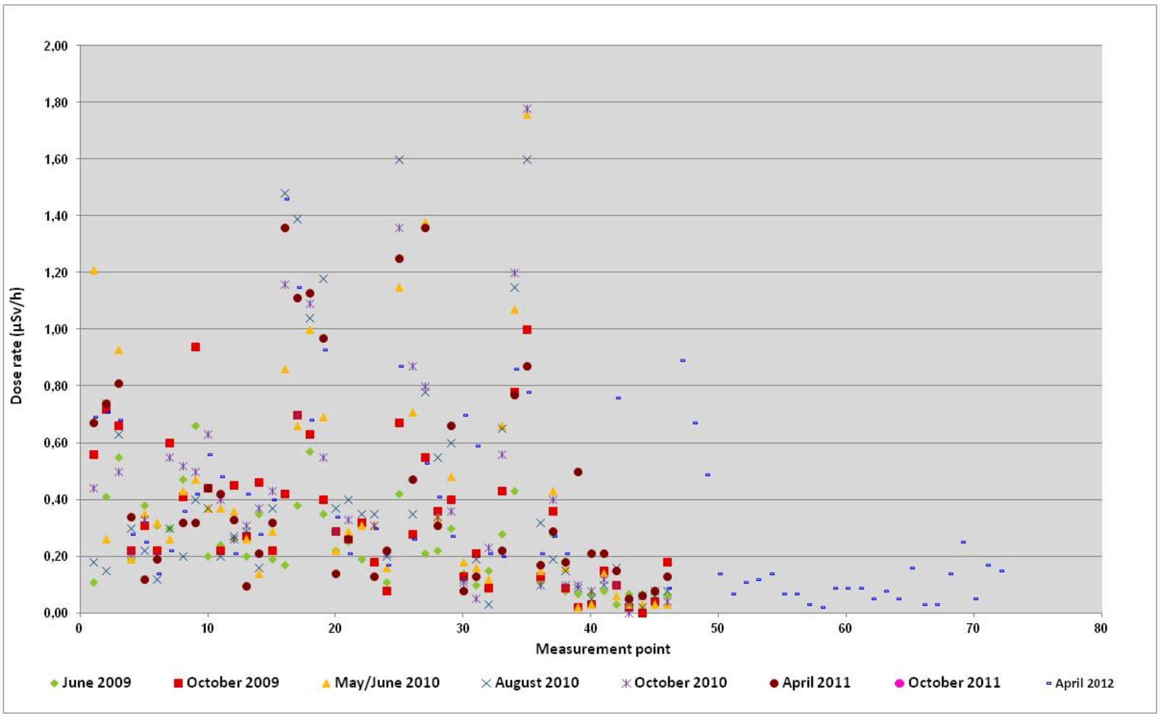

Figure 4: Evolution of ambient dose rate measurements on GPK2 platform

Concerning ambient measurements, they range between the background noise level, up to 0.66 µSv/h, with an average of 0.24 µSv/h.

It can be observed on figures 3 and 4 that in October 2009 after 7 months of circulation the dose rates values increased. The maximum contact value now reached 11.93 µSv/h, with an average of 1.49 µSv/h (Guéry, 2009; Maquet, 2011). The same behaviour is observed for the ambient measurements with a maximum of 1.00 µSv/h and a mean of 0.35 µSv/h. Table 1 presents the cumulative volume of circulated geothermal fluid and the corresponding contact dose rate average for each survey (Maquet, 2011; Orsat, 2012a).

The third survey in May/June 2010 took place after around 6 months of circulation. In between the circulation tests of 2009 and 2010, a general cleaning of the installation was performed. Here the maximum contact value is 11.26 µSv/h and the average is 1.60 µSv/h (Cuenot et al., 2010a; Maquet 2011). Those values are close to those of the preceding survey, done after almost the same circulation duration. Maximum ambient value reached 1.73 µSv/h, for an average of 0.43 µSv/h. During the next two surveys performed in August and October 2010, the contact dose rate values continued to increase (Cuenot et al., 2010b). The maximum observed dose rate value reached 17.50 µSv/h in October 2010 after 11 months of continuous circulation. The average contact dose rate was 1.78 µSv/h in August 2010 and increased to 2.23 µSv/h in October 2010 (Table 1). However the ambient dose rate average values kept rather stable at 0.43 and 0.44 µSv/h, in August and October 2010 respectively. The results show that the general level of dose rate tends to increase as a function of the circulation time and cumulative volume of geothermal fluid that circulated in the surface installations.

Table 1: Circulated volume and dose rate average for each survey

Survey	Volume (m^3)	Contact dose rate average value (µSv/h)
June 2009	308 500	0.89
October 2009	404 000	1.49
May/June 2010	297 600	1.60
August 2010	408 300	1.78
October 2010	494 000	2.23
April 2011	160 000	1.88
October 2011	132 000	1.86
April 2012	112 800	1.77

In April 2011, despite a lower circulated volume and a cleaning of the surface installation, the contact measurements are still rather high, with a maximum of 9.85 µSv/h and an average of 1.88 µSv/h (Table 1). It would mean that either the mechanical cleaning by high-pressure water jetting is not efficient enough or, as the measurements were done 5 days after the end of the test, radioactivity continued to increase even after the end of circulation. The latter hypothesis is related to the origin of radioactivity in the installation that will be discussed in chapter 3 below. The ambient measurements remains stable, with an average value of 0.44 µSv/h.

The survey of July 2011 was carried out in order to characterize the effect of a cleaning of surface installation performed in June 2011 (Maquet, 2011). The results show that, for 60% of the measurements points, the dose rate level remained unchanged after cleaning and that the dose rate average reached 2.16 µSv/h. Maquet (2011) assumes that the cleaning does not remove the radioactive particles, but only transfers them to other parts of the installation.

In October 2011, a few days before the end of the circulation test, another survey was performed, which shows dose rates values close to the April 2011 survey: maximum dose rate is 9.70 µSv/h and the average is 1.86 µSv/h (Table 1), for a circulated volume also similar to April 2011. The mean value for the ambient measurements is again 0.44 µSv/h.

Finally the last survey was carried out in April 2012, after only 2 months of circulation and a lower circulated volume of geothermal fluid. The maximum dose rate value reached 9.49 µSv/h and the average was 1.77 µSv/h (Orsat, 2012a).

On figure 3 we can observe 5 peaks showing the highest dose rate values. The measurements points have been identified and correspond to specific places on the surface installation, that are:

- 1) Filters on reinjection lines
- 2) Part of reinjection line
- 3) Reinjection line to GPK3
- 4) Part of reinjection line
- 5) Reinjection pumps

Thus it looks like that the highest levels of radioactivity are not randomly located on the installation, but occur on specific places. This observation is confirmed when looking at the precise mapping of dose rate values on the installation: figures 5 and 6 present the cartography of dose rate values on GPK2 installation. Figure 5 correspond to the first survey performed in June 2009 and figure 6 to the survey of October 2010, which shows the highest values. The comparison of both figures highlights the fact that the general level of dose rate increased between both surveys, because of the increase of the circulated fluid volume. Moreover it is clearly visible that most of the highest values (in brown, red and orange colour) are located on the right of figure 6. All this part of the surface installation corresponds to the reinjection line. It is in agreement with the observation made on figure 3, where the 5 peaks also correspond to equipments on reinjection line. On figure 6, the other part showing high values is located on the left, at the outlet of heat exchangers. On the contrary, on the production lines (centre of figures 5 and 6), the dose rate values are rather low.

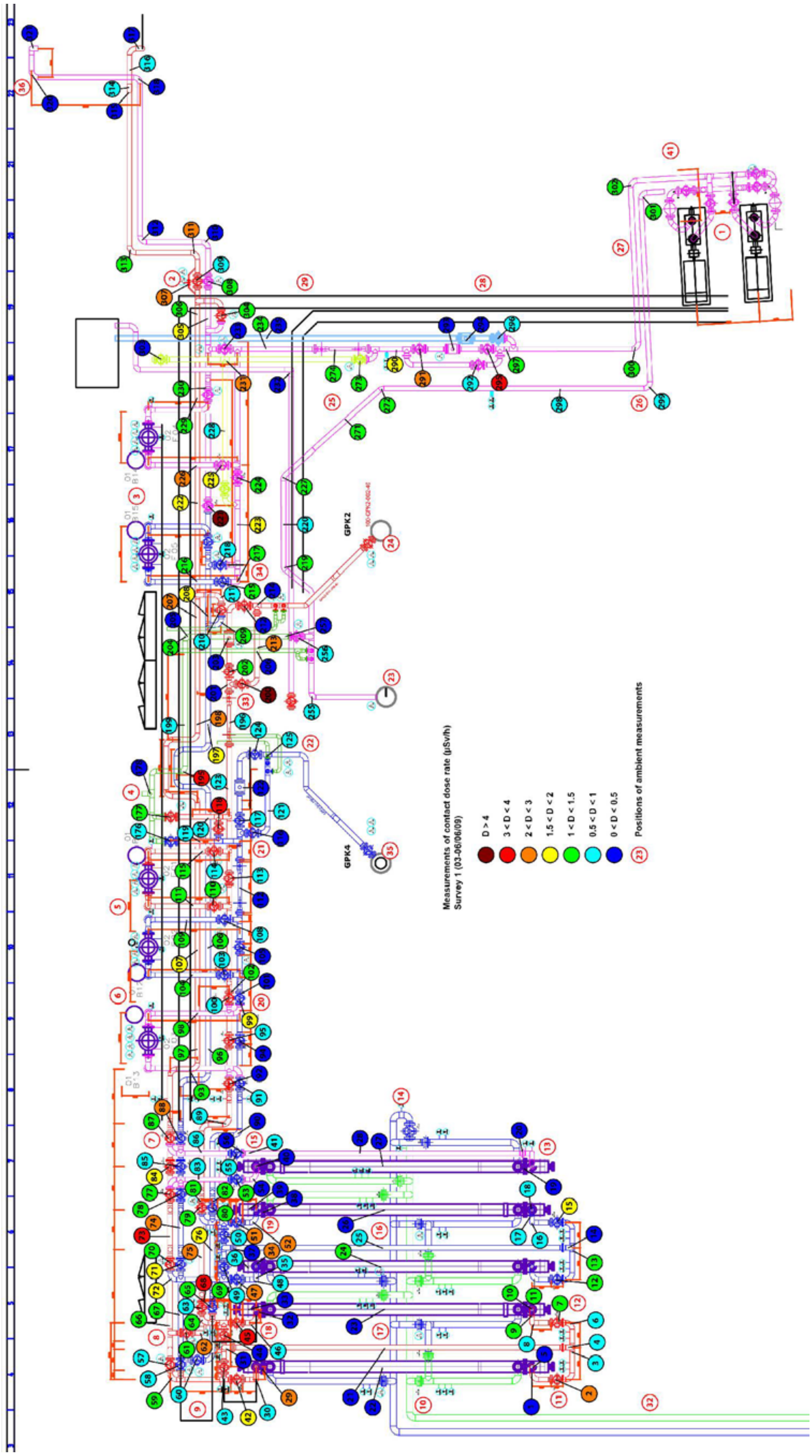

Figure 5: Cartography of dose rates on GPK2 platform from the survey performed in June 2009. The coloured circles correspond to the contact measurements points and the colour code is function of the dose rate value. The red circles with a red number indicate the ambient measurement positions.

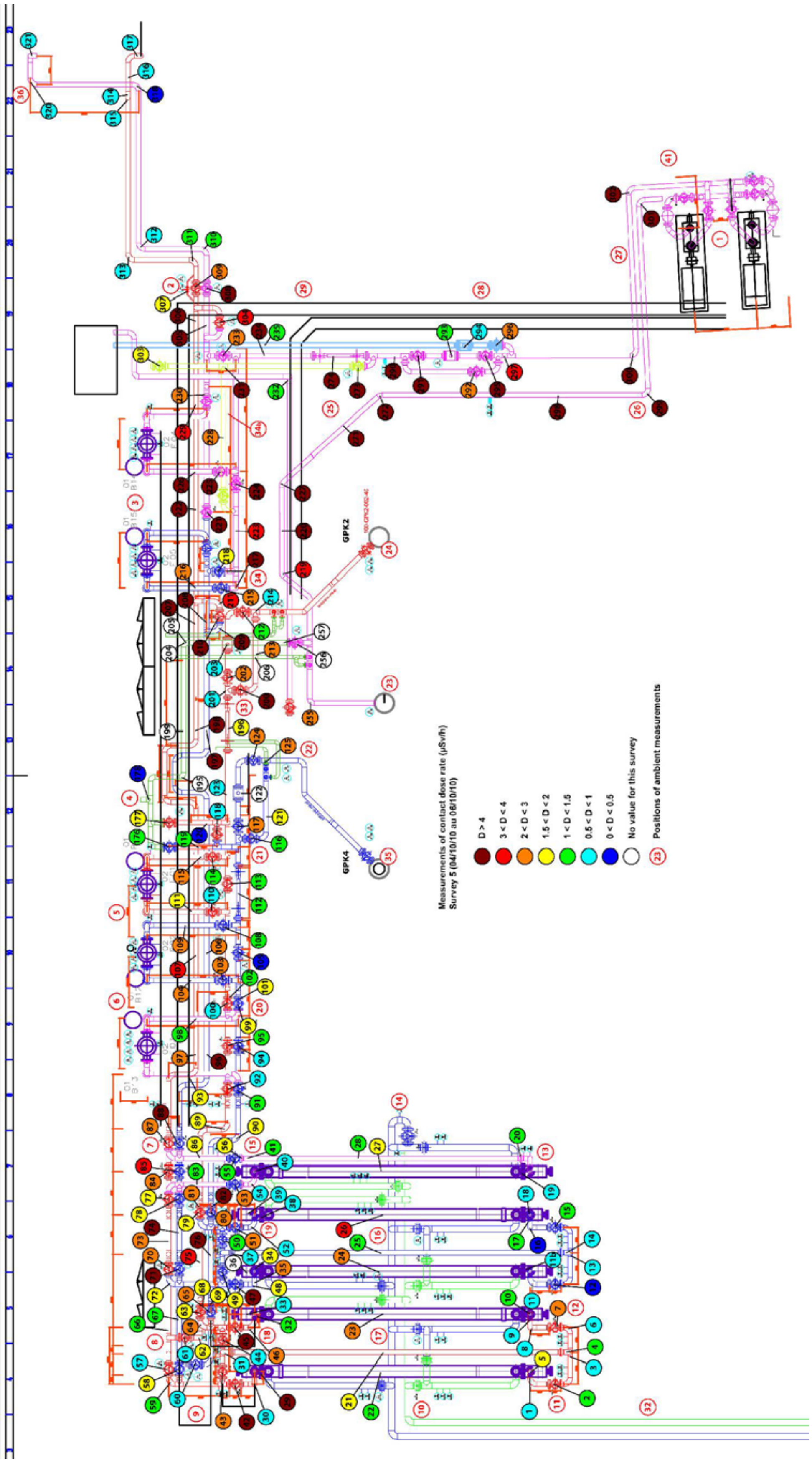

Figure 6: Cartography of dose rates on GPK2 platform from the survey performed in October 2010. The coloured circles correspond to the contact measurements points and the colour code is function of the dose rate value. The red circles with a red number indicate the ambient measurement positions.

From the results obtained on GPK2 installation, the main conclusions are the following:

- The general level of dose rate tend to increase as a function of the circulated volume
- The highest dose rate values are observed mainly on the equipments that belong to the reinjection line, where the temperature is lower (around 70°C)
- The mechanical cleaning by high-pressure water jetting seems not efficient enough to remove all radioactive particles

2.3 Results on GPK1 platform

The above results are confirmed with the observation made on GPK1 installation. In 2009, GPK1 has been progressively used for reinjection with low reinjected volumes. In the circulation tests of 2010, 2011 and 2012, on the contrary, the volume of geothermal fluid injected into this borehole increased more and more. Thus, in October 2009, at the end of the 2009 circulation test, the contact dose rate average value was only 0.14 µSv/h and the ambient 0.07 µSv/h. At the end of the 2010 circulation test, the same values increased to 2.28 µSv/h and 0.33 µSv/h respectively. In April 2011 and October 2011, at the end of both 2011 circulation tests, the contact dose rate mean values reached 2.26 µSv/h and 2.78 µSv/h. The ambient averages were 0.28 µSv/h and 0.31 µSv/h. Finally in April 2012, most of the fluid was reinjected into GPK1 and as a consequence, the contact and ambient dose values increased to 2.86 µSv/h and 0.61 µSv/h respectively. The observations made on GPK1 installation confirm the conclusions from GPK2 platform.

3. ORIGIN OF RADIOACTIVITY IN THE GEOTHERMAL INSTALLATION

3.1 Scaling in the surface installation

From the precise mapping of dose rate values on GPK1 and GPK2 surface installation, it appears that the highest levels are located mainly on reinjection lines, where the fluid temperature is lower (60-70°C). Moreover, when dismantling some parts of the installation (pipes, heat exchangers, filters), it has been observed that scaling occurs inside the installation. Figure 7 shows a part of a pipe dismantled from the reinjection line. A clear black deposit is visible inside this pipe.

Mineralogical analysis of the scales has been performed and revealed that the main dominant species are sulfates (solid solutions of Barite, $BaSO_4$ and Celestine, $SrSO_4$) and sulfides (Galena, PbS). On figure 7, the black deposits correspond to Barite and Celestine. For both of them, their saturation index in water tend to increase with temperature of about 50-70°C inducing their precipitation. The above temperatures correspond to those found in the reinjection line of the geothermal installation, which explains the presence of these scales in the installation.

Figure 7: Pipe removed from the reinjection line and covered with black deposits

3.2 Scaling in the boreholes

Moreover, the same process has been observed in the boreholes through a series of logging measurements. A first gamma-ray (GR in the following) logging series was carried out and revealed a huge increase of the gamma-ray values in respect to former measurements. For example, Figure 8 shows a comparison of gamma-ray logs performed in GPK1 in May 2005 and December 2011. The GR values from December 2011 are rather low in the near surface (depth interval where variations of water level occur), but at around 70 m depth, a remarkable peak is observed (around 13 000 gAPI). It corresponds to an increase by a factor 170 in respect to May 2005 values (Orsat, 2012b). It is associated to the fact that GPK1 has been more and more used for reinjection.

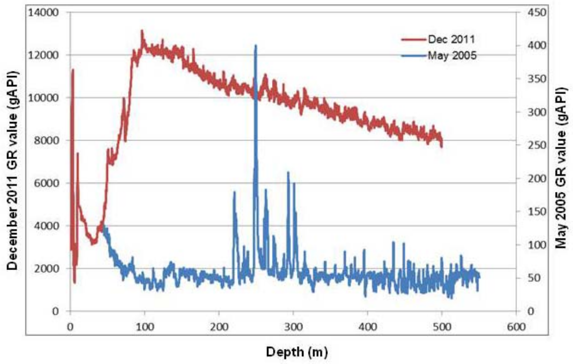

Figure 8: Gamma-ray logging in GPK1. In red, December 2011 data; in blue, May 2005 data.

Then, the GR values tend to decrease as a function of the depth, but still remain at a high level at 500 m depth. A second series of logging measurements was performed afterwards, involving a video camera logging survey and a caliper survey (measurement of borehole internal casing diameter). Figure 9 shows a picture taken with the camera in borehole GPK1 where the presence of scales is visible on the casing wall at around 80 m depth. Moreover, the caliper survey revealed a slight diminution of the casing diameter in respect to its nominal value. This means that scaling can reached a thickness of a few millimeters.

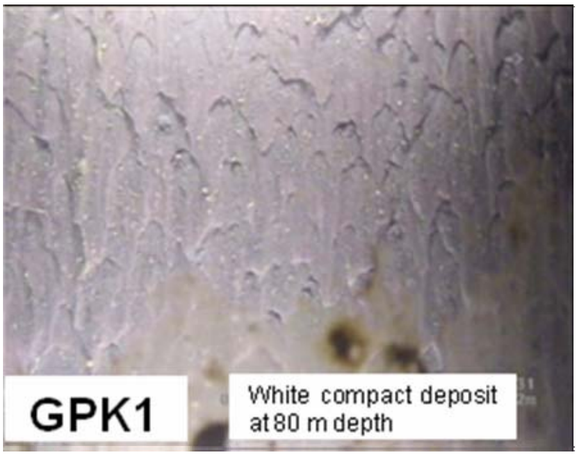

Figure 9: Camera view of GPK1 borehole wall showing the presence of deposits.

GR and caliper logging surveys were also performed in reinjection borehole GPK3 in December 2012; they show the same results as GPK1 survey, namely, high GR values and presence of a few millimeters thick scales.

However, the same series of logging measurements was performed into production borehole GPK2. They show slightly increased GR values and the presence of very localized scaling. In GPK4, mainly used as a production well, a very low level of radioactivity and very few deposits were observed in the December 2012 logging surveys. So the level of GR values and the volume of scales are very different in the production boreholes than in reinjection boreholes.

Thus, from the observation made on surface installation and in the geothermal boreholes, a clear relationship appears between the presence of scaling and the occurrence of radioactivity.

3.3 Relationship scaling/radioactivity: origin of radioactivity

A research work has been carried out to understand the origin of radioactivity linked with scaling formation. Eggeling et al. (2013) showed that, during the precipitation of Barite and Celestine, atoms of Barium (Br^{2+}) and Strontium (Sr^{2+}) can be replaced by atoms of Radium (Ra^{2+}), because of their similar atomic radius. Then, ^{226}Ra can be incorporated in the Barite and Celestine crystals. ^{226}Ra is a product of the disintegration of Uranium ^{238}U, that is naturally present in the granite.

Concerning Galena (PbS), the process which leads to the occurrence of radioactivity is the incorporation of a radioactive isotope of lead, namely ^{210}Pb, during the crystallization of Galena.

The radioactive material which is indeed created is due to natural chemical processes, but they are not the results of a dedicated industrial process. Thus in the nuclear regulation, they are called **NORM** (Naturally Occurring Radioactive Material).

4. RADIOPROTECTION

4.1 Some values for comparison

The general dose rate level is not very high, especially the ambient dose rate values, which best represent work positions. For instance, ambient values of 1 µSv/h would require for a worker to spend 1000 hours in this environment, in order to reach the legal limit of 1 mSv/h over a period of 12 consecutive months. This is clearly unrealistic. Moreover, to give a better idea of the level of dose rate on the surface installation, it is interesting to compare it with examples of human natural and medical exposure (Table 2). For example, in France, the average natural dose received per year is 2.6 mSv, coming mostly from exposure to radon but also cosmic and telluric radiations. This value is higher than the legal limit of 1 mSv.

Table 2: Examples of received dose

Origin of radiation	Dose received
Cosmic radiations	0.31 mSv/year (average in France)
Telluric radiations	0.60 mSv/year (average in France)
Exposure to Radon	1.5 mSv/year (average in France)
Flight Paris-New York	0.02 mSv
Chest X-ray	0.05 mSv
Chest scanner	5.7 mSv
Abdominal scanner	12 mSv

4.2 Radioprotection procedures

In regards to the values in Table 2 and considering the annual working time, the dose rates observed in the surface installation are rather low. Nevertheless, because of the occurrence of radiations, even low, the French nuclear regulation requires to set up radioprotection procedure for the workers, but also for the public as the Soultz site regularly welcomes visitors. Two “PCR” (in French: Personne Compétente en Radioprotection, Person skilled in radioprotection) have been trained and nominated in the GEIE and are in charge of the organization of radioprotection in the company and exchanges with authorities.

Installation zoning

The first measure was to set up a zoning of the installation, in respect to the French regulation, which implies a restricted access. A blue zone, called “protected area” has been defined to ensure that, outside this zone, no one would receive a dose of 1 mSv over 12 consecutive months. Practically, its limits are defined by an average dose rate of 0.5 µSv/h, which is in the range of the average ambient dose rates measured on surface installation. A blue line has been drawn on the floor and blue radioactive signs have been installed (Figure 10).

Figure 10: Blue line and radioactive sign defining the protected area

A second zone has been defined around the place where residues of scales from filters, heat exchangers and pipes are stored. Due to their radioactive content, these residues have to be stored into barrels, in a specific place that is isolated from the main installation. Due to the accumulation of radioactive material inside this area, which involves an increase of the dose rate, a green zone, called “controlled area” has been defined, where the access is even more restricted than in the “protected area” (Figure 11).

Figure 11: Storage of radioactive residues in the controlled area

<u>Radioprotection procedures for workers</u>

Some of the works that have to be done on the installation require specific radioprotection procedures for the workers.

The first is imposed by the French regulation and is linked with the zoning of the installation: in protected or controlled areas, workers must wear a personal dosimeter (Figure 12) which records the cumulative dose that they received during their working time. Every 3 months, they are sent back to IRSN (Institut de Radioprotection et de Sûreté Nucléaire). Here they are analyzed to see if the legal dose of 1 mSv over 12 consecutive months has been reached or not. Results are sent to the works doctor in charge of the company, who has to proceed to a specific follow up of the workers. Moreover, if external workers need to enter in the protected area, a work permit is required and must be signed by the PCR.

Figure 12: Personal dosimeters of GEIE’s employees

A detailed analysis of every work station has to be made by the PCR so as to evaluate the dose that can be received. This has to be further confirmed by real measurements. Once it has been done, specific radioprotection procedures can be set up. In view of the low dose rate values and the emitted radiation (mostly α and β, few γ), the risk of external contamination is very low. So adapted equipment must be worn by workers (one-use suits, gloves, glasses). Masks are important to wear, because there is a risk of internal contamination in case of ingesting or inhaling particles that can emit α and β radiations, which are very dangerous for health if present inside bodies. These equipments are especially important when the job involves a possible direct contact with radioactive material (pipe dismantling, filters and heat exchangers opening and cleaning, Figure 13).

Figure 13: Equipment worn for opening a heat exchanger

<u>Radioactive material management</u>

As the residues (mainly scales and other rock particles) shows non negligible dose rate values, they have to be stored in a specific place on the installation (described above), but their removal from the site must follow the French nuclear regulation. Thus an agreement with ANDRA (French national agency for radioactive waste management) was established. When the blue barrels (visible on figure 11) are full, ANDRA comes on site for removing the wastes, after a full radioactive characterization that has to be performed by an approved body. On the ANDRA storage site, the wastes are treated as low activity material, either for elimination or long-term storage.

5. CONCLUSIONS

The occurrence of NORM on geothermal sites which exploits a reservoir located in rocks naturally containing radionuclides implies many efforts to manage all issues related to radioactivity. In France, the regulation for nuclear safety is well-established and very strict. However the existing regulation about NORM mainly concerns mining industry, oil and gas industry and underground water exploitation, but is not very adapted to geothermal energy. It means that the laws have to be interpreted to fit with the regulation. But with the development of geothermal energy in France and in Europe, it is very likely that a specific regulation will quickly emerge.

However, preventing the occurrence of radioactivity in the surface installation would avoid all the issues described in this paper. As simple cleaning seems not to be effective enough, a scientific research has been carried out about scaling inhibitors (see paper of Scheiber et al., 2013; Scheiber et al., 2012). As scaling and occurrence of radioactivity are closely related, preventing the formation of scales could be a way to decrease or, in the best case, to completely avoid the radioactivity.

The relationship between increasing levels of radioactivity and formation of sulfate/sulfide scaling is clearly established. But, other processes may be involved and contribute to the occurrence of radioactivity: contamination of metallic surfaces of equipments (pipes, heat exchangers, filters, pumps) or incorporation of radionuclides in other precipitates (oxides for instance). Thus, further measurements and research works are still needed and will be carried out at the Soultz site, but also probably at other geothermal power plants in the Upper Rhine Graben.

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