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The success story of TUNA and the FOPH in Switzerland – a report of Dr. Philipp Steinmann

In the aftermath of the nuclear accident of Fukushima Daiichi, in 2012, the Federal Office of Public Health FOPH began planning an automated measuring network to monitor the river water below the nuclear power plants. Meanwhile TUNA detectors are installed at five different sites.

Dr. Philipp Steinmann, Deputy Head of Section of the FOPH, reports on his experiences with TUNA detectors and Scienta Envinet's monitoring networks in the following article (published in "AQUA & GAS" N° 10/2015).

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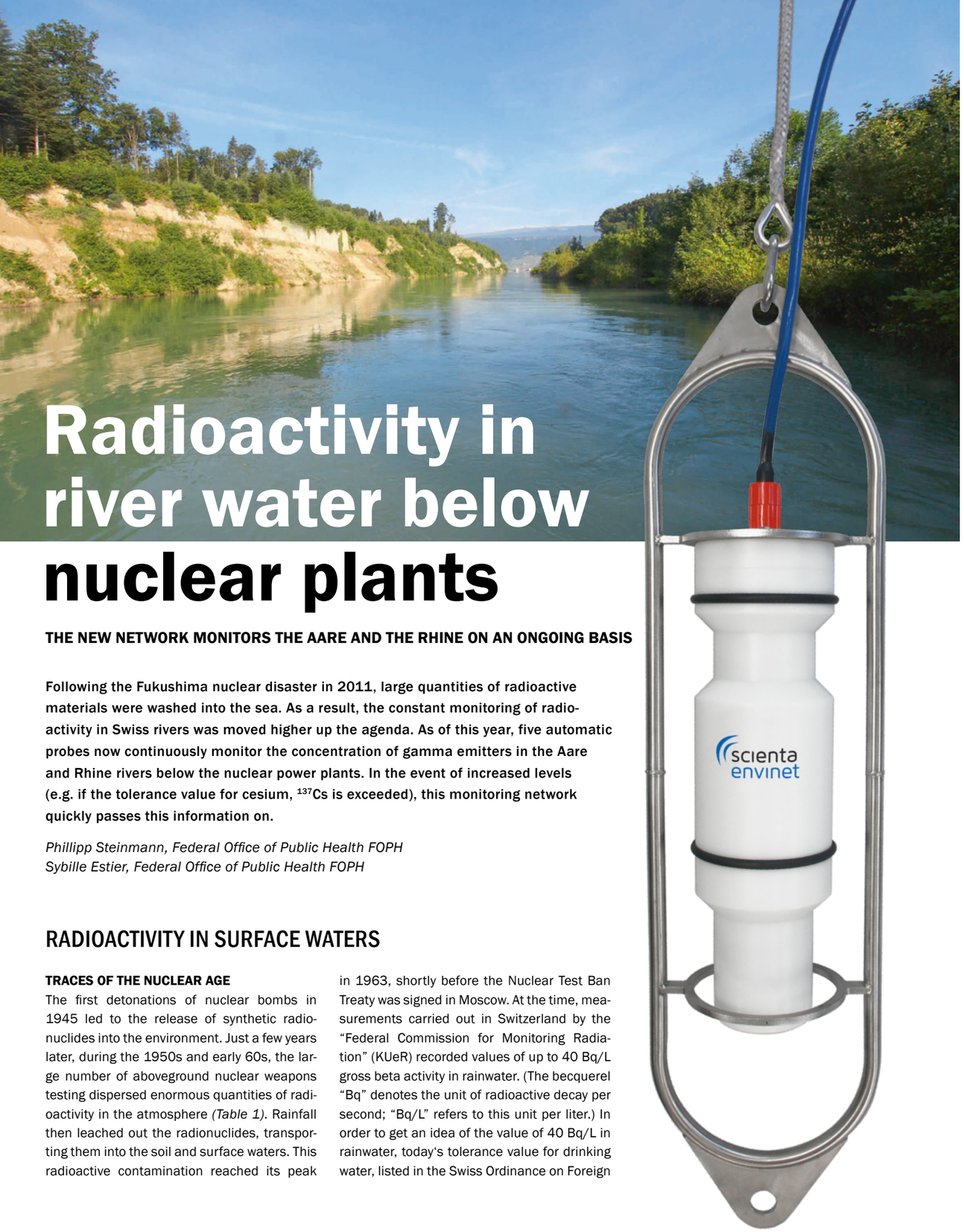

Radioactivity in river water below nuclear plants

THE NEW NETWORK MONITORS THE AARE AND THE RHINE ON AN ONGOING BASIS

Following the Fukushima nuclear disaster in 2011, large quantities of radioactive materials were washed into the sea. As a result, the constant monitoring of radioactivity in Swiss rivers was moved higher up the agenda. As of this year, five automatic probes now continuously monitor the concentration of gamma emitters in the Aare and Rhine rivers below the nuclear power plants. In the event of increased levels (e.g. if the tolerance value for cesium, ^{137}Cs is exceeded), this monitoring network quickly passes this information on.

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RADIOACTIVITY IN SURFACE WATERS

TRACES OF THE NUCLEAR AGE

The first detonations of nuclear bombs in 1945 led to the release of synthetic radionuclides into the environment. Just a few years later, during the 1950s and early 60s, the large number of aboveground nuclear weapons testing dispersed enormous quantities of radioactivity in the atmosphere *(Table 1)*. Rainfall then leached out the radionuclides, transporting them into the soil and surface waters. This radioactive contamination reached its peak in 1963, shortly before the Nuclear Test Ban Treaty was signed in Moscow. At the time, measurements carried out in Switzerland by the "Federal Commission for Monitoring Radiation" (KUeR) recorded values of up to 40 Bq/L gross beta activity in rainwater. (The becquerel "Bq" denotes the unit of radioactive decay per second; "Bq/L" refers to this unit per liter.) In order to get an idea of the value of 40 Bq/L in rainwater, today's tolerance value for drinking water, listed in the Swiss Ordinance on Foreign Substances and Constituents in Foods (OSEC) as 1 to 10 Bq/L for beta emitters, may be used as a basis. A large percentage of the gross beta activity applied to relatively short-lived fission products like ^{95}Zr or ^{144}Ce with a half-life of 65 and 280 days respectively. Today, these radionuclides can no longer be detected in the environment. Frequently used as the reference nuclide for the radioactive pollution of the environment and still traceable in environmental samples today, cesium-137 (^{137}Cs, half-life of 30 years) accounted for approximately 2 Bq/L rainwater in 1963. Tritium (^{3}H) is not included in the gross beta activity. Its activity in rainwater in 1963 reached 300 Bq/L (the mean level in western Europe). As tritium is less radiotoxic, this level is well below the tolerance value of 1000 Bq/L. When Switzerland started monitoring tritium activity in 1966, the first readings indicated a maximum of 120 Bq/L tritium in rain (with a mean of approximately 40 Bq/L). Furthermore, the gross beta activity in rain had already decreased by a factor of more than 10 by 1966 (maximum of 2.5 Bq/L, mean of almost 1 Bq/L). As the majority of radioactive materials are retained in soil, the radioactivity level detected in the rivers was invariably below that of rain. For 1963, the KUeR reported gross beta activities in river water of between 0.5 and 1 Bq/L. Today, the occasional traces of artificial radioactivity in river water samples amount to a mere one thousandth of this level *(Table 2)*.

From the 1970s onwards, the issue of discharges from the newly erected nuclear power plants (NPP) into surface waters became an issue and had to be included in the monitoring network. In particular, the contaminated material disposed of in the Irish Sea by the nuclear fuel reprocessing and decommissioning site at Sellafield hit the headlines.

In 1986, the Chernobyl reactor disaster led to the contamination of large areas of Europe, particularly due to ^{137}Cs. There was a significant increase in ^{137}Cs activity in soil and foodstuffs, as well as in surface waters. Up to 10 Bq/L ^{137}Cs were detected in some samples of drinking water in Switzerland, with reservoir water reading up to several 100 Bq/L. In September 1986, five months after the accident, ^{137}Cs activity in Lake Lugano was approximately 1 Bq/L.

FUKUSHIMA AND SWITZERLAND

In spring 2011, 25 years after Chernobyl, the tsunami triggered by the great Tohoku earthquake destroyed the nuclear reactors of the Fukushima Daiichi nuclear power plant complex. This led to large quantities of radioactivity being released into the sea along with the discharge of the water used for cooling and for extinguishing the fire. Extremely high ^{137}Cs concentrations of up to 68,000 Bq/L were measured in the seawater in the immediate vicinity of the releases [2]. Although, due to the strong dilution, these concentrations dissipated rapidly and no problematic concentrations were detected in offshore waters [2], the events in Japan raised awareness of the possibility of a contamination of surface waters in the wake of a nuclear disaster. In the aftermath of Fukushima,as in many other countries, Switzerland reviewed and revised its emergency

From 1952	As a result of aboveground nuclear tests, artificial radioactivity enters the atmosphere and surface waters.
1956	The Federal Commission for Monitoring Radiation (KUeR) is founded in Switzerland and begins analysing samples of river water
As of 1970	Nuclear power plants are put into operation
1986	Nuclear accident in Chernobyl; large areas of Europe are contaminated
From 1990	Eawag carries out specific studies into the activity of synthetic radionuclides from nuclear power plants in the Aare and Rhine rivers [1]; monthly measurements begin in the trace range (mBq/L)
March 2011	Fukushima Daiichi nuclear disaster, releasing large quantities of radioactivity into the sea [2]
2012	Following discussions with SVGW and ENSI, the FOPH plans to establish an automatic monitoring network for river water using NaI scintillators. Publications with scenarios of "Fukushima discharges" in Switzerland [3,4]
2013	Political calls for action regarding radioactivity in surface waters [5]; Swiss Federal Council ratifies an automatic monitoring network (May); the first NaI probe is installed in Basel (June)
2014	Publications on the dispersion of discharges from the nuclear power plants in the rivers [6] and the impact on the drinking water supply [7]; NaI probes are installed in Niederried and Aarau (June)
2015	Probes are commissioned in Laufenburg (January) and near Hagneck (June). Daily readings of the NaI river monitoring probes are published on www.radenviro.ch (under "URAnet aqua" monitoring network, from Nov. 2015)

Table 1 Some of the milestones in establishing the automatic monitoring of radioactivity in river water

[mBq/l]	Rainwater	River water (Hagneck)	Groundwater	Drinking water (Biel treatment plant)
^{40}K (nat)	–	1–30	< 2	–
^{7}Be (nat)	100–3300	2–25	< 1	< 2
^{210}Pb (nat)	10–140	< 1–40	< 10	< 4
^{3}H (nat/syn)	1 000–60 000	1 000–10 000	–	–
^{137}Cs (syn)	< 100 (–600)	< 0,1–3	< 0,3	< 0,4
^{60}Co (syn)	–	< 0,1–3	< 0,2	< 0,4
^{54}Mn (syn)	–	< 0,1–1	–	< 0,4
^{65}Zn (syn)	–	< 1	< 0,3	< 1
$^{239/240}$Pu (syn)	–	0,003–0,009	–	–

Table 2 Concentrations and concentration ranges in mBq/L of natural (nat) and synthetic (syn) radionuclides, which have been recorded in the last few years in various types of water samples (1 mBq/L = 0.001 Bq/L) *(Fotos: FOPH and ENVINET GmbH)*

scenarios and contingency plans. Particular attention was paid to the contamination of surface waters and drinking water, partly as a result of a study on the potential consequences of an accident in a Swiss nuclear power plant, with similar discharges of radioactive material as in a block of the Fukushima Daiichi NPP [3]. In this context, and following discussion with the Swiss Federal Nuclear Safety Inspectorate ENSI and the Swiss Gas and Water Industry Association SVGW, in 2012, the Federal Office of Public Health FOPH began planning an automated measuring network to monitor the river water below the nuclear power plants. Suitable monitoring probes had come onto the market in recent years; therefore, the technical basis had already been established. The undertaking was supported by a number of political calls for action and a commission from the Federal Council *(Table 1)*.

CURRENT LEVELS OF RADIOACTIVITY IN SURFACE WATERS

Essentially, the current level of radioactivity in Swiss surface waters originates from four main sources:

- Leaching of radioisotopes from the atmosphere via rainwater
- Leaching of radioisotopes from the soil
- (Authorized) discharges from nuclear power plants, research facilities and companies
- Medical discharges

The first group is made up of cosmogenic ^{7}Be, i.e. created by cosmic rays in the upper atmosphere, and the likewise naturally occurring radon progenies (including ^{210}Pb).
The nuclides of the natural decay series of uranium and thorium may be washed out of the soil, together with older deposits (i.e. particularly ^{137}Cs, ^{90}Sr and traces of plutonium). Besides ^{137}Cs, ^{90}Sr and tritium, the liquid discharges from nuclear installations, the PSI and the interim storage facility ZWILAG primarily contain activation products like ^{58}Co, ^{60}Co and ^{54}Mn. The companies and incineration plants mainly produce tritium and ^{14}C. Nuclides like ^{131}I, ^{90}Y, ^{177}Lu and the new ^{223}Ra are the result of medical applications. As shown in *Table 2*, the natural radioisotopes ^{210}Pb, ^{40}K, ^{7}Be and tritium, the majority of which occur naturally nowadays, are predominant in river water. For the most part, synthetic radionuclides only occur in very small activities of less than mBq/L (0.001 Bq/L).

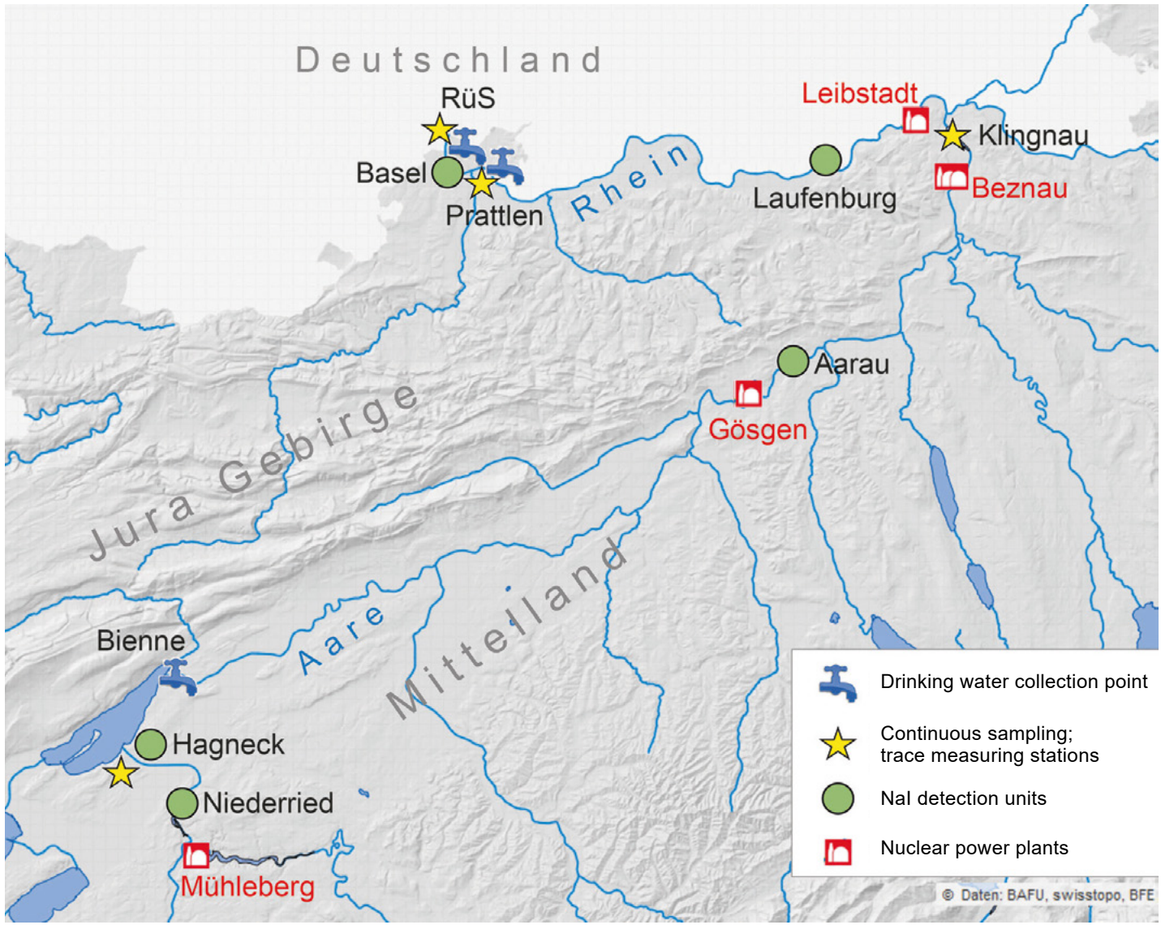

Figure 1 Sites of the NaI detectors; Swiss Federal Institute of Aquatic Science and Technology (Eawag) stations with continuous sampling for trace measurement (Hagneck, Klingnau and Pratteln); the Rhine monitoring station RüS Weil a.Rh. (measurements carried out in the Cantonal Laboratory Basel City); drinking water collection points in Lake Biel and the Rhine (Pratteln and Basel)

MONITORING SURFACE WATERS

The current monitoring network of radioactivity in precipitation, surface waters and groundwater comprises 21 rainwater collectors (11 of which measure the gamma nuclides + tritium, 10 tritium only), 16 sample stations for river water (11 gamma + tritium, 5 tritium only) and four groundwater collection points. Besides water samples, sediments, aquatic plants and fish are also studied. The Eawag trace measuring stations of Hagneck, Klingnau and Pratteln form an essential part of the system of monitoring measures *(Figure 1)*. The stations collect river water in proportion to the outflow on an ongoing basis. Every month, the collected water is concentrated (hydroxide precipitation) and then subjected to a gamma-spectroscopic analysis. This is the only method of obtaining sufficiently low detection thresholds to be able to record the current low concentrations and calculate the monthly levels. Another crucial collection point is the Rhine monitoring station at Weil am Rhein, where the Cantonal Laboratory Basel City supervises the radioactivity measurements. In addition to the abovementioned partners, the FOPH also collaborates with the PSI, CHUV-IRA (Lausanne), the University of Geneva and Suva to monitor the surface waters. As of this year, five additional automatic, continuously measuring NaI detection units have been integrated in the monitoring network *(Figure 1)*.

AUTOMATIC MONITORING OF THE RIVERS USING NaI DETECTORS

TASKS PERFORMED BY THE AUTOMATIC NAI DETECTION UNITS

The new, automatic monitoring probes are designed to rapidly detect significantly elevated levels of radioactivity in river water. The detection limit of approximately 1 Bq/L enables a lower reporting threshold to be set in the range of the OSEC tolerance value, which is 10 Bq/L for ^{137}Cs. In accordance with the OSEC limit for drinking water, a second threshold has been determined, namely 1000 Bq/L for ^{137}Cs, which triggers an alarm. If the lower reporting threshold is exceeded, the text messaging service notifies the responsible FOPH office, which then verifies the readings, identifies the cause of a possible contamination and forwards the information. If the second threshold is exceeded, the National Emergency Operations Centre is alerted immediately, which initiates the appropriate action.
Even the exceedance of tolerance values are detected and reported, thereby enabling the necessary steps to be taken before there is any risk to public health as a result of radionuclides in drinking water. Nevertheless, as the authorized discharges have been specified to ensure compliance with the tolerance value, it is unlikely that this will ever be exceeded during the day-to-day operation of nuclear power plants. Therefore, the NaI detection units are not designed to replace the Eawag's monthly measurements: these are even able to document minimal discharges from nuclear installations due to their detection threshold, which is more than a thousand times lower.

MEASURING NETWORK

In light of the original discussions with the SVGW, it made sense to install the first probe in the Rhine or, to be more precise, in the data processing center of the Industrial Werke Basel (IWB) at the sampling point where the waters of the Rhine are used to produce drinking water in the Langen Erlen area. The first installation confirmed that the chosen technology met the specified objectives and that adequate monitoring was in fact feasible

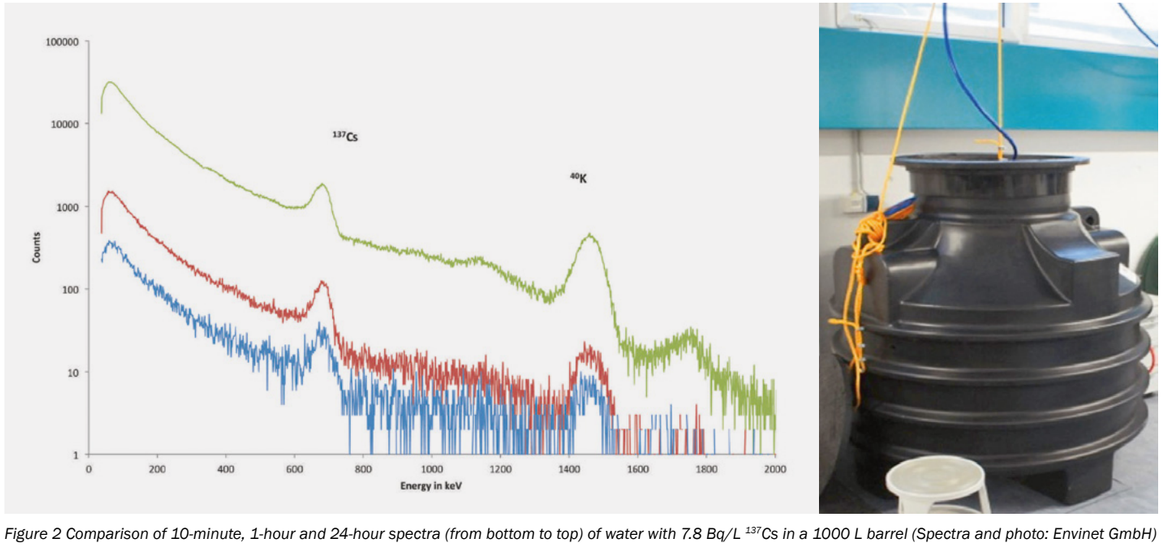

Figure 2 Comparison of 10-minute, 1-hour and 24-hour spectra (from bottom to top) of water with 7.8 Bq/L ^{137}Cs in a 1000 L barrel (Spectra and photo: Envinet GmbH)

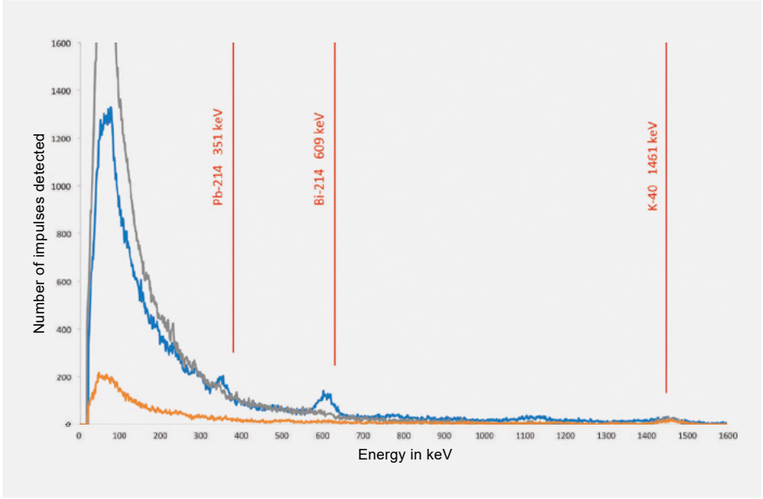

Figure 3 Comparison of 1-h spectra at low water, probe A (orange curve), high water in the fixed probe A (blue curve) and high water in a freely suspended probe B (gray curve). Probe A detects significantly more radon progenies in high water (peaks of ^{214}Pb and Bi). In the case of B, the entire subsurface is elevated at high water as the probe is floating (see text)

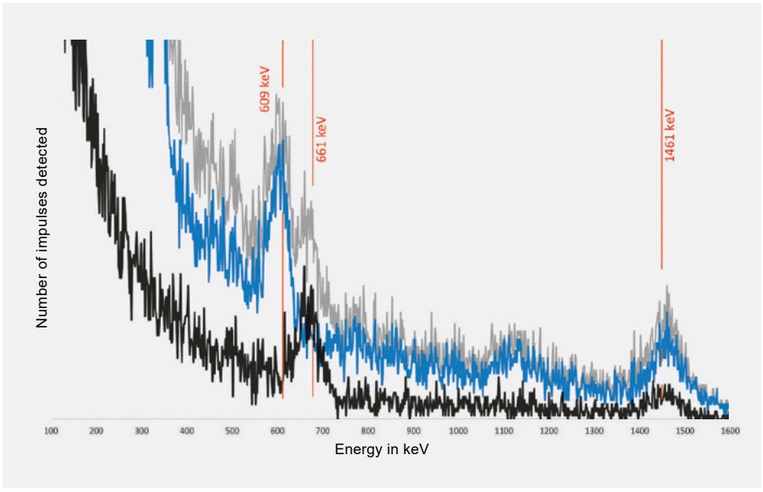

Figure 4 Comparison of 1-h spectra in Aare water at high water (blue curve) in water with 7.8 Bq/L ^{137}Cs in a 1000 L calibration tank (black curve, spectrum courtesy of ENVINET GmbH). The resolution of the NaI detector enables a clear differentiation between the 661 keV peaks of ^{137}Cs and the 609 keV peaks of the radon progeny ^{214}Bi. The sum of the two spectra (gray curve) illustrates that 7.8 Bq/L ^{137}Cs could even be detected in the elevated subsurface of high water. The 1461 keV peak is due to naturally occurring ^{40}K (Spectra: FOPH)

at reasonable expense. The other detection units were thus to be positioned accordingly so as to monitor the water below the nuclear power plants. The FOPH deemed a distance of 5 to 20 km downriver from the NPP to be appropriate for measuring the water independently of NPP operators in a manner that was representative for each section of the river. At this distance, river water is largely mixed, and yet there is still sufficient time to inform the drinking water plants that are located further down the river if the water sample exceeds the recommended levels.

The search for potential locations focused in particular on the waters upstream from retaining weirs, where there is a relatively constant water level and some sites are not exposed to strong currents. Thanks to the active support of the companies enlisted (BKW Energie AG, IBAarau Kraftwerk AG, Energiedienst Holding AG Laufenburg), three suitable locations were quickly identified. Niederried Station is situated 5 km downstream of Mühleberg NPP but above wnburg Station is 14 km below Leibstadt NPP and 27 km downriver of Beznau NPP, but above the drinking water collection points in Pratteln and Basel. Hagneck Station, which was established 18 km below Mühleberg NPP in cooperation with the Federal Office for the Environment FOEN and the Federal Institute of Metrology METAS, ensures redundancy for Niederried Station while allowing a direct comparison of the Eawag's trace radioactivity measurements and the FOEN's hydrological measurements under one roof.

MEASURING TECHNOLOGY

The deployed probes contain 3" NaI (sodium iodide) detectors, which have a ‚visibility range' in water of around one meter. The NaI crystals register the gamma radiation and the probe records an energy spectrum (compare *Figure 2–4*). The energy resolution of the NaI spectra is considerably coarser than the spectra of the germanium detectors used in the laboratory. It is nonetheless sufficient to differentiate between natural and synthetic radionuclides. In addition to the natural nuclides (^{40}K, radon progenies ^{214}Pb/^{214}Bi, thoron progeny ^{212}Pb), the probes' configuration also enables them to detect synthetic radionuclides. The synthetic radionuclides captured by the probe include nuclear fission products (^{137}Cs, ^{134}Cs, ^{131}I, ^{132}Te, ^{140}La, ^{103}Ru, ^{99}Mo) as well as the activation products caused by neutron radiation (^{54}Mn, ^{58}Co, ^{60}Co, ^{65}Zn). The probes record each of the spectra over a period of ten minutes; they are then analysed by the embedded PC and forwarded to the monitoring center. Once a full hour has elapsed, the six 10-minute spectra are added together and re-evaluated, which allows for a lower detection threshold (1 Bq/L for ^{137}Cs). A slightly better detection threshold is obtained in the daily analyses (a total of 24 hour spectra) *(Figure 3)*. A small amount of a potassium salt is incorporated in the detector for energy calibration, therefore the ^{40}K peak in the energy spectrum is present in all spectra. The 10-minute spectra often do not present any other peaks. However, several peaks of the natural nuclides ^{214}Bi und ^{214}Pb are clearly visible in most of the hour spectra, particularly at high water when a large number of radon progenies enter the river *(Fig. 3)*. *Figure 4* studies the impact of increased natural radiation on the detection threshold for synthetic nuclides, e.g. ^{137}Cs. A comparison of the spectra shows that it is still possible to definitively detect a concentration of slightly below the tolerance value, despite a high contribution of natural radiation. In addition to the activities of the individual nuclides in "Bq/L" (decays per second per liter), the probe also calculates the dose rate from the spectrum in "nSv/h" (nanosievert per hour) – a unit that expresses radiation intensity at the measuring point, from which the radiation dose can be derived.

INSTALLATION AND MAINTENANCE

Due to its "visibility range", the detector unit should ideally be surrounded by at least a meter of water on all sides. To this end, the probe is lowered into the water on a cable, which is attached to the top edge of the frame. A second (steel) cable is fixed to the bottom of the hoist, looped through a ring below the probe and thereafter extends back up to the surface of the water *(Figure 5)*. Loosening the lower steel cable enables the detector unit to be pulled up on the upper cable for maintenance. The probe can then be repositioned by tautening the lower cable again. If there is no lower cable, a heavier flow pushes the probe towards the surface, which leads to an increase in the recorded dose rate. On the other hand, the risk of being carried away by large pieces of floating debris is much lower for a floating probe than if it is permanently installed. Therefore, care was taken to place a pole upstream of the probes that are fixed on both sides to protect them from floating debris. A blue undersea cable between 20 to 30 meters long connects the detection units to a weatherproof box containing a GPRS modem and a battery in case of power outages. The data monitoring center retrieves the readings and status reports at regular intervals via modem. Data transfer takes place via the *SwissMetNet* automatic monitoring network hosted by MeteoSchweiz.

Maintenance work includes cleaning every six months and an annual constancy test using a screw-on reference source (^{137}Cs).

Figure 5 A) Plan for installing a water probe on an underwater structure;
B) A hoist mounted on the riverbank;
C) The lower anchoring system
The hoist holds the blue connector cable and the supporting steel cable in place underwater.
The lower steel cable is used to position and secure the probe
(Image A: D. Wyder, Metas; Image C: taf-taucharbeiten ag)

EXPERIENCE TO DATE

As stated, apart from the peaks of ^{40}K (the potassium salt incorporated in the detector for calibration) and ^{214}Pb/^{214}Bi (radon progenies), no other peaks are visible in the spectra provided by the monitoring probes under normal conditions. In other words, the individual radionuclides are below the detection threshold. By contrast, the dose rate can be specified at all times. Therefore, for normal operation, the chronological sequence of the dose rate is the best indication of the fluctuations and anomalies in the readings. The most striking reading is the extremely small dose rate: at a mere 2 nSv/h, it is low compared to the local dose rate of approximately 100 nSv/h, commonly recorded 1 m aboveground in the Central Plateau. This is because the surrounding water serves as an effective screen, shielding the detector from cosmic and terrestrial radiation. Brief spikes (increases) of the dose rate recorded in the river water may become apparent in lengthy series of measurements. These are almost invariably linked to heavy rainfall and thunderstorms *(Figure 6)*. An analysis of the spectra shows that, in these situations, it is primarily the peaks of the radon progenies that rise sharply, as illustrated in *Figures 2* and *3*. Torrential rain washes them out of the soil (and also directly from the air) into the rivers. It cannot be determined with absolute certainty whether radon itself is also discharged into the river water from the soil, lingering there long enough to add to the signal. Moreover, several of the dose rate ‚storm peaks' also display an increased ^{40}K peak, like the thunderstorm on 11 August 2014 in the Canton of Bern *(Figure 6)*. This can be

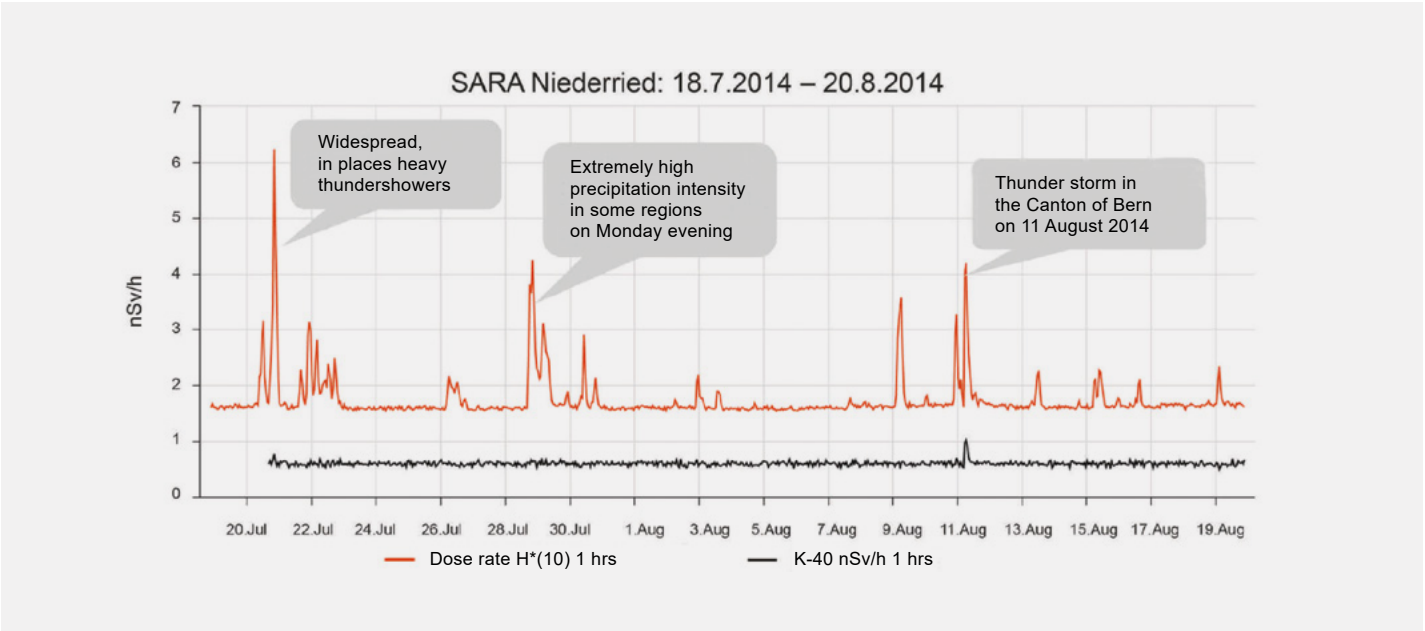

Figure 6 Time series for the dose rate and K (here in units of nSv/h) measured at Niederried Station (Spectra: FOPH)

accounted for by vast quantities of water with a high concentration of suspended particles, including potassium-rich clay minerals. A probe that is freely suspended in water indicates a special "high water increase" in the dose rate. As the probe floats upwards at high tide, it is shielded by a thinner layer of water. Therefore, a larger contribution of cosmic rays increases the radiation reading. This explanation is supported by the observation that, although this probe recorded spectra with a higher overall level at high water, they do not show any significant peaks for radon progenies *(Figure 3)*. In this case, therefore, the radon progenies are not the cause of the signal increase. To date, the NaI probes have not detected any synthetic radionuclides.

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