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Water Rock Interaction [WRI 14]

A review of possible origins of the uranium “plume” in the aquifer under the EPIC site in the Chernobyl Exclusion Zone

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Abstract

The uniqueness of the Chernobyl accident lies in the fact that so much radioactive material was discharged to the atmosphere as solid fuel particles from the reactor core. Between the 26th of April and the 6th of May 1986 more than 6 tons of small particles of highly radioactive uranium oxide fuel were discharged to the atmosphere and were responsible for more than 75 % of the radioactive contamination on the ground in the exclusion zone. In 1987, about 800 trenches had been dug in the exclusion zone to prevent re-suspension and to protect workers from contamination. In 1999, the IRSN, in collaboration with IGS and UIAR, equipped trench 22 (CPS) in order to monitor radionuclide migration in the environment (water, soil, plants). At the EPIC site high uranium concentrations were observed in the groundwater downstream from trench 22. We discuss the possible origins of this uranium “plume”.

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1. Introduction

25 years have passed since the Chernobyl NPP accident (26/04/1986), but up to now scientists are still working on answering the question “what are the consequences of the accident in Ukraine?”; more specifically the question about the radionuclide (RN) migration processes taking place today and in the future. Since 1999, the French Institute of Radioprotection and Nuclear Safety (IRSN), in collaboration with the Ukrainian Institute of Agricultural Radiology (UIAR/NUBiP) and the Ukrainian Institute of GeoSciences (IGS), has been studying the impact of radionuclides migration from the contaminated waste trench 22. This trench is located 2.5 km South-West from the Chernobyl NPP and is perpendicular to the general flow direction. The Experimental Platform in Chernobyl (EPIC), which includes trench 22 (Figure 1), was equipped with the necessary facilities for radioecological and hydrogeological *in situ* investigations.

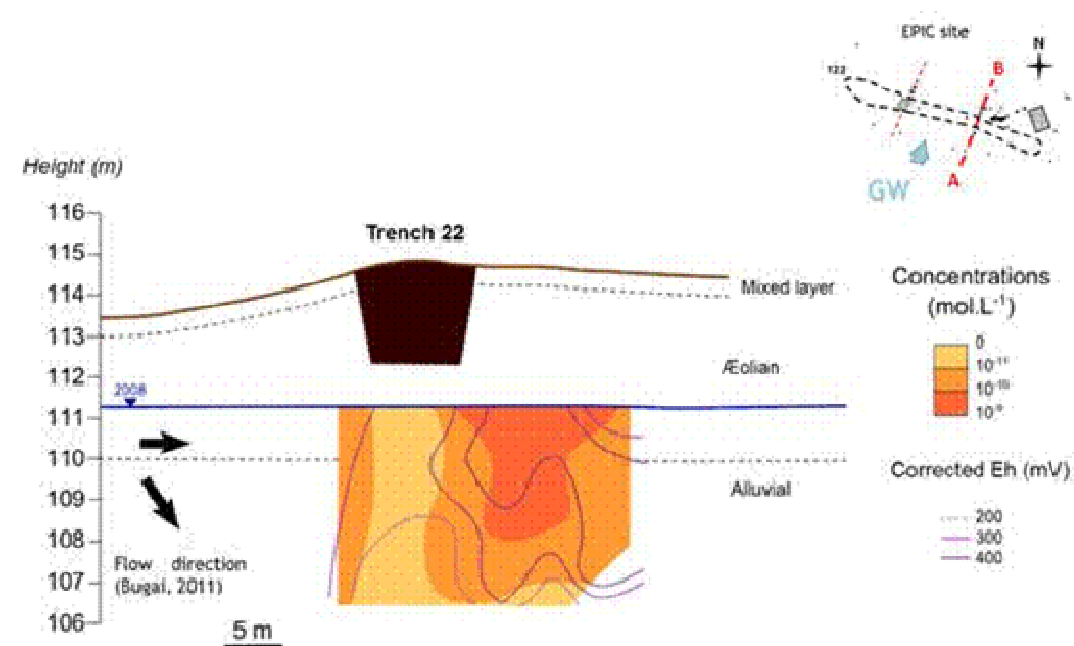


Figure 1: map of the EPIC site included trench 22 and ^{238}U concentrations and corrected Eh interpolated profile in October 2008 [1]. Uranium-238 concentrations are ranging between $1.7 \cdot 10^{-11}$ (sampled in the 1-98-3 piezometer in October 2008) and $5.7 \cdot 10^{-9} \text{ mol.L}^{-1}$ (sampled in the 1-06-1 piezometer in October 2008). U was analysed by ICP-MS, detection limit: $2.1 \cdot 10^{-11} \text{ mol/L}$ – 25 samples. The used Eh cell was a Sentix ORP (WTW), values were corrected by a uniform correction of 216 (for temperatures between 10 and 15 °C).

Following the Chernobyl accident, clean-up activities were carried out in order to prevent fires at the “Red Forest” (i. e., pine trees killed by radiation) and to lower radiation levels at the ground surface to protect workers. The decontamination resulted in the removal of the upper 10-15 cm thick soil layer (contaminated topsoil layers containing fuel particles) and demolition of vegetation and buildings within about an 8 km² area. The radioactive waste resulting from this clean-up was disposed locally in trenches and/or artificial mounds directly into the local sandy soil without any use of impermeable liners [2].

Relatively high uranium concentrations were observed down-gradient from trench 22 (Figure 1). Through this study, the possible origins of this uranium plume were investigated.

2. Uranium possible origins

2.1 *Is the dissolution of fuel particles the main origin for uranium?*

Two main types of fuel particles having different chemical behaviour were identified in the material buried in trench 22 [3]:

- U-O particles, more or less oxidized as UO_2 or UO_{2+x}
- U-Zr-O particles probably formed following the fusion of the reactor's core, resulting from the interaction between nuclear fuel and the combustible sheath in zircolay Zr-Nb.

Since the burial, fuel particles have been weathered and the associated radionuclides have been released in the environment [3]. The dissolution rate of the fuel particles depends on their specific surface (which depends on size and oxidation state), soil solution and soil pH [3]. A kinetic fuel particle dissolution model was run using trench 22 conditions; it showed that most oxidized particles such as UO_{2+x} were a priori dissolved 25 years after the burial and that all radionuclides linked to dissolved fuel particles should have been released into the soil solution. Therefore, currently mainly the less degradable compounds (UO_2 and ZrU_3O_x) should remain, and uranium should not be released anymore.

Some correlation between Eh and uranium distribution in groundwater is observed (figure 1), which is consistent with the fact that uranium is sensitive to redox changes. More reduced conditions are associated with low U concentrations just below the trench while in more oxidizing conditions higher U concentrations are observed down-gradient.

Because of the ^{235}U enrichment of the fuel particles, an isotopic approach seemed suitable to characterize the origin of U. According to the estimations, fuel particles were 1.1 % enriched in ^{235}U content [4]. Analyses on the soil-leachate solutions obtained by concentrated hydrochloric acid gave ratios for $^{238}\text{U}/^{235}\text{U}$ between 40 and 125 [4]. If the uranium downstream from the trench originated from the fuel particles, similar isotopic ratio should be measured. Uranium isotopic ratios were studied and measured by TIMS [1]. The measured ratios in the aquifer ($^{238}\text{U}/^{235}\text{U}$ from 136.2 ± 2.2 to 140.0 ± 2.2) are close to the natural ratio ($^{238}\text{U}/^{235}\text{U}$: 137.88) [1], which suggests that the origin of the uranium in groundwater may not be the FP (fuel particles) dissolution, so the question remains: What is the origin of the high U concentrations downstream from the trench and where is the uranium from fuel particle dissolution in groundwater? Could the trench conditions have contributed to the mobilisation of natural U in the groundwater?

We here put forward 3 hypotheses to explain the lack of change in the $^{238}\text{U}/^{235}\text{U}$ ratio in groundwater: first, natural leaching by rainwater, with a pH around 5-5.5, is not acid enough to leach uranium from fuel particles; second, it could be that the uranium of the dissolved oxidized particles have been released but are trapped in the soil, finally uranium could have moved out of the investigated profile under another form (velocity in the aeolian layer: 10 m/y). Potential processes for the immobilization of U in the trench or soil is discussed below.

2.2 *Potential trapping of uranium released from fuel particles dissolution*

Uranium is known to have a great affinity with organic matter. Material buried in the trench contains a non-negligible amount of organic matter. This organic matter comes mostly from tree trunks, forest contaminated soils and plant litter (organic matter: 3-6 %), plant debris (like needles, branches, etc.) or other plants (such as herbs, shrubs, etc.). The kinetics of organic decaying ranges from slow decaying of less degradable compounds ($T_{1/2}$ = 7-42 years) for coarser materials to fast decaying of easily degradable compounds ($T_{1/2}$ = 3-4 years) for fine litter. 25 years after burial, most of the organic compounds easily

degradable should have been transformed, hence mainly the less degradable compounds should persist in the trench (ex.: trunks). Fuel U is likely to be absorbed on secondary organic material (humus), it seems that there is still such secondary organic material in the trench.

In contrast the decomposition of organic matter can affect the chemical conditions of soil and may affect the migration of uranium through the unsaturated zone. The precipitation (rain, snow) may also modify the chemical conditions. The variation of pH for example, can result in the leaching of radionuclides and cations in solution as uranium can be sorbed on different minerals.

Further more likely reduced conditions in the trench would efficiently prevent U migration toward the groundwater. In contrast, leaching of the bottom of the trench likely to raise the Eh values, when the water table rises, may be likely to mobilize U toward the groundwater.

The presence of soil microorganisms may also affect the migration of uranium as uranium can be removed by microorganisms. Iron reducing bacteria (FRB) and sulphate reducing bacteria (SRB) can reduce enzymatically U(VI). The latter plays the role of electron acceptor involved in organic matter oxidation [5]. Studies showed that the removal of uranium is largely due to the microbiological activity in the first centimetres of sediment below the sediment-water interface. Deeper, where microbial activity is less intense, the main uranium removal process seems to be abiotic.

After this review, it is clear that the origin of uranium high concentrations down-gradient is not due to U release by dissolution of fuel particles, but might be linked to the combination of different processes. A better characterization of the bacterial community in the trench along with U concentration and composition in and directly below the trench for instance could help in understanding the processes that prevent the transfer of uranium towards the aquifer.

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