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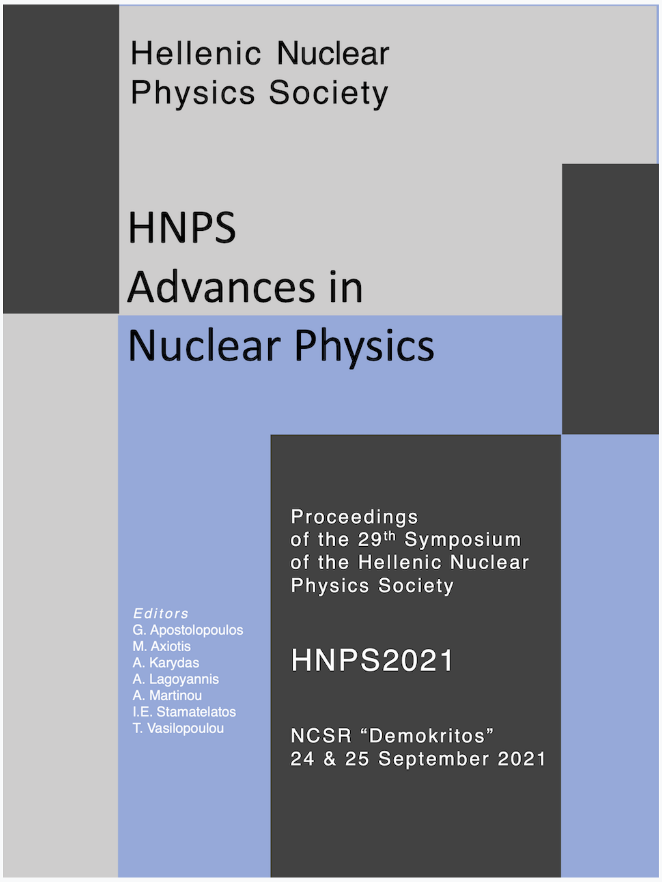

Radiation exposure of microorganisms living in radioactive mineral springs

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Abstract The TIRAMISU collaboration gathers expertise from biologists, physicists, radiochemists and geologists within the Zone-Atelier Territoires Uranifères (ZATU) in France to study the response of microorganisms living in naturally radioactive mineral springs. Mineral springs are isolated ecosystems, extremely stable over geological time scales, which display different physicochemical and radiological parameters compared to their surroundings. Water and sediment samples collected in 27 mineral springs of the volcanic Auvergne region (Massif Central, France) have been analyzed for their microbial biodiversity and radionuclide content. Among the microorganisms present, diatoms (a microalgae species), widely used as environmental bioindicators of water quality, have shown to display an exceptional abundance of teratogenic forms in the most radioactive springs studied (radon activity up to 3700 Bq/L). The current work presents a first assessment of the dose received by the diatoms inhabiting naturally radioactive mineral springs. The radiological risk for microorganisms living in freshwater environments was estimated using the ERICA tool. Most of the sampled mineral springs were highly above the risk threshold of 10 μGy/h due to the large concentrations of radium in the sediments (up to 50 Bq/g). The complete radiological data on water and sediments are used as inputs to Monte Carlo simulations at micro- (GATE) and nano- (Geant4-DNA) scale in order to assess the direct and indirect damages on the diatom DNA.

Keywords natural radioactivity, mineral springs, dose, simulation, diatoms

INTRODUCTION

Naturally radioactive mineral springs can be described as small waterbodies of various physicochemical properties and elevated levels of radionuclide content in comparison to their

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surroundings, in which minerals and radioisotopes of the three natural decays series (^{238}U, ^{232}Th and ^{235}U) are abundant [1–2]. Specifically, ^{226}Ra ($t_{1/2}$ = 1600 y) and its gaseous descendant ^{222}Rn ($t_{1/2}$ = 3.82 d), both emitting alpha particles of maximum energy 4.8 MeV and 5.5 MeV respectively, are found in high concentrations in the sediment and the water. A correlation between mutation rates and radioactivity levels has already been observed for organisms inhabiting naturally radioactive environments [3].

Auvergne, situated in Massif Central of France, is a volcanic region rich in mineral springs and high uranium content [4]. Diatoms, unicellular photosynthetic microorganisms, thrive in these peculiar environments [5], while recent studies indicate exceptional abundance of teratogenic forms in the most radioactive springs in Auvergne [6].

Diatoms account for a great part of the carbon dioxide fixation in the aquatic environments. Their dimensions are of the order of a few micrometres and they are uniquely characterized by their frustule, a silicate membrane serving as a skeleton [7] which remains as fossil after their death [8]. In addition to being used as water quality bio-indicators due to their sensitivity and direct response to environmental stresses [9], diatoms are now studied for their response to natural radioactivity in the frame of the TIRAMISU project.

The TIRAMISU (biodiversiTy in the RAdioactive Mineral Springs in Auvergne) project is dedicated to the study of the radiological and radiotoxic effects of natural radioactivity on microorganisms living in naturally radioactive mineral springs within ZATU [10]. In the current work, we are focusing on the radiological content of these ecosystems and the simulation of the radiation exposure on diatoms.

For the characterisation of the ecosystems, campaigns of sampling have been launched including the collection of water, diatoms, sediment, and travertine cores, in 27 locations. The radiological content of the water and sediment samples was analysed using γ -spectroscopy. The study and calculation of the radiation effects on diatoms has been feasible due to Monte Carlo Simulation Codes (MCSC). Especially at the nanoscale, the stochastic nature of the interactions responsible for the DNA damage due to ionizing radiation, make the use of MCSC an essential part of radiological studies [11].

In this study, a risk assessment based on the measured specific activity values of the radionuclides in each mineral spring, was initially performed for the biota of freshwater ecosystems using the ERICA tool. Microdosimetric calculations followed using GATE, while nanodosimetry was covered by the use of Geant4-DNA.

MATERIALS AND METHODS

Sampling and Analysis

Water samples have been collected from 27 mineral springs in Auvergne, Massif Central, France (*Fig*). A High-Purity Germanium (HPGe) well-type detector was used for γ-spectroscopic analysis at LPC (Clermont Ferrand, France). For the determination of the ^{222}Rn activity, the 352 keV γ-ray line of ^{214}Pb ($t_{1/2}$ = 27.06 min) was considered under the assumption of secular equilibrium.

Sediment samples were also collected from the different sites. The measurements for the determination of ^{226}Ra activity were conducted at least four weeks after the sealing, allowing for the secular equilibrium between ^{222}Rn and ^{226}Ra to be reached. These analyses were conducted at Subatech (Nantes, France) using a coaxial HPGe detector. The data have been then used as an input to the simulations as described in the next section.

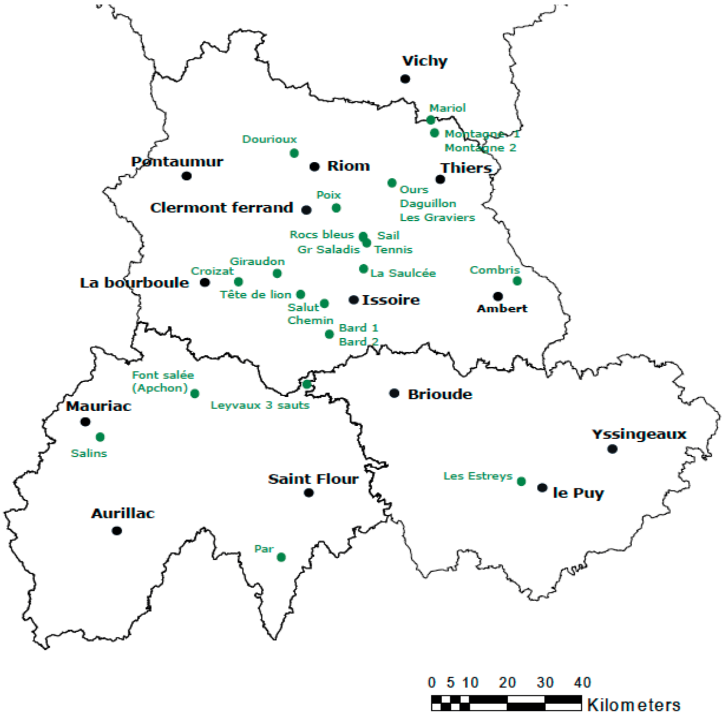

Fig. 1. *Map of the sampled mineral springs, represented in green, in Auvergne*

Simulation

ERICA tool v.1.3 [12] is based on datasets of radiological factors in order to evaluate the radiological risk to biota. In the current study, a risk evaluation (eq. 1) for phytoplankton in freshwater ecosystems was conducted for each individual mineral spring using the measured sediment activity concentrations (in Bq/kg) for ^{226}Ra, ^{238}U and ^{210}Pb. The ^{222}Rn water activity concentrations were excluded due to absence of noble gazes from the radiological factor database of ERICA. The risk assessment is performed using eq. 1, ,where DR in μGy/h is the estimation of the total weighted absorbed dose rates according to ERICA tool and SDR (= 10 μGy/h) is the default dose rate screening value for the protection of the ecosystems [13].

$$RQ = \frac{DR}{SDR} \qquad (1)$$

For the the need of a complete evaluation of the radiation exposure, MCSC were used. Geant4 is an opensource simulation toolkit for the tracking of particles through matter [14], offering the ability of modelling and simulating from nanoscale up to macroscale [15-16].

GATE, an opensource platform based on Geant4 libraries initially dedicated to medical physics applications [17–19], has been employed for the microdosimetric simulation of the absorbed dose to diatoms due to the natural radioactivity present in the mineral springs.

In the current work, we modelled the diatom as a 20 μm radius water sphere surrounded by 100 μm radius water and/or sediment sphere serving as the surrounding environment. When considering sediments, a mixture consisting of a user-defined “dry sediments” material and “G4_WATER” was used, characterized by its porosity values (in %) as described in eq.2:

$$Porosity = \frac{Volume_{water}}{Volume_{sediments}} * 100 \qquad (2)$$

Focusing on the two most abundant radionuclides measured in the water and the sediment of the mineral springs, the alpha spectra of ^{222}Rn and ^{226}Ra were introduced as volumetric, isotropic sources surrounding the diatom and irradiating it either externally and/or internally.

For the needs of the Geant4-DNA simulation, information on the type, kinetic energy and direction of the particles entering the diatom were also recorded in a file using the dedicated GATE "PhaseSpace" (PhS) actor.

Geant4-DNA is an opensource MCSC dedicated to micro- and nano-dosimetry allowing the tracking of particles down to eV energies [20]. It allows the evaluation of Single and Double Strand Breaks (SSBs and DSBs respectively) due to the direct energy deposition of the ionizing particles as well as the simulation of the indirect DNA damage due to water radiolysis [21].

In this study, Geant4-DNA was employed for the assessment of the SSBs and DSBs using the clustering algorithm DBSCAN [22].

The diatom, in this case, consists of a water sphere of 10 μm radius with a spherical nucleus of 0.5 μm, while the source introduced here is the PhS file collected from the GATE simulation as described before. We collected the specific energy (z in Gy) deposited in cylinders of two different sizes: 10 nm diameter and 5 nm height representing a nucleosome and 2 nm diameter and 2 nm height representing 10 DNA base pairs.

The radiation-induced DNA SSBs and DSBs due to the direct energy depositions were evaluated using the DBscan clustering algorithm. The formation of SSBs is considered a function of the energy deposition following a probability distribution function which is zero for deposited energy (E_{dep}) < 5 eV and reaches unity for $E_{dep} \geq 37.5$ eV. The minimum number of SSBs to form a DSB is set to 2 within a radius of 3.3 nm, representing roughly the distance between 10 DNA base pairs.

The water radiolysis and the subsequent interactions of the reactive species have been simulated in the dedicated Geant4-DNA chemistry module. The source is again the PhS file collected from the GATE simulation, consisting of the alpha particles entering the nucleus, while the collection of the events is taking place in a 1 x 1 x 1 μm^3 water box. The radiochemical yield (G-value) of reactive species due to the water radiolysis is described by eq.3:

$$G(t) = \frac{N(t) \times 100}{E_{dep}(eV)} \qquad (3)$$

where N(t) is the number of molecules of a given radiolytic species as a function of time t.

RESULTS AND DISCUSSION

γ-Spectroscopy

The measured ^{222}Rn concentration activities (A_{Rn} in Bq/L) in the waters of the mineral springs range between 1.3 and 4000 Bq/L. The sampled locations were categorized according to the values of ^{222}Rn activities in three classes of low (A_{Rn} < 100 Bq/L), medium ($100 \leq A_{Rn} < 1000$ Bq/L) and high ($A_{Rn} \geq 1000$ Bq/L) activity levels. In overall, 58% of the mineral springs belong to the low activity level class while 11% belong to the high activity level class. Among all the locations, the highest values of Rn activity concentration in the water (A_{Rn} = 4000 Bq/L) are measured in La Montagne.

Concerning the sediments, ^{210}Pb is present in 80% of the mineral springs with a mean specific activity value of 1.4 Bq/g. ^{238}U and ^{226}Ra are detected in the sediments of all the mineral springs with specific activities ranging between 7.6 10^{-3} and 0.5 Bq/g, and 0.028 to 52 Bq/g, respectively.

Simulation

The Risk Assessment for the phytoplankton by ERICA, using the activity concentration values in the sediments measured by γ- spectroscopy in the mineral springs, indicated that there is a very low risk due to the presence of ^{238}U and ^{210}Pb in the sediments. On the other hand, it was found that there is a high risk for the phytoplankton in 71% of the sampled locations due to the presence of ^{226}Ra in the sediments.

Using GATE, the dose rate (μGy/h) received by the diatoms as a function of the porosity of the sediments was calculated (*Fig. **1***). The specific activities and activity concentrations used here represent mean real values that were measured from the samples. At the x-axis, 0% porosity represents a totally dry sediment composition (0% of water), while in the case of 100% porosity we are referring to a composition of 100% water.

The observed trend is that the dose rate received by the diatoms decreases as a function of the porosity. This indicates that the contribution of ^{226}Ra (present in the sediments) to the dose received by the microorganisms living in mineral springs is higher in comparison to the dose received due to the presence of ^{222}Rn in the water.

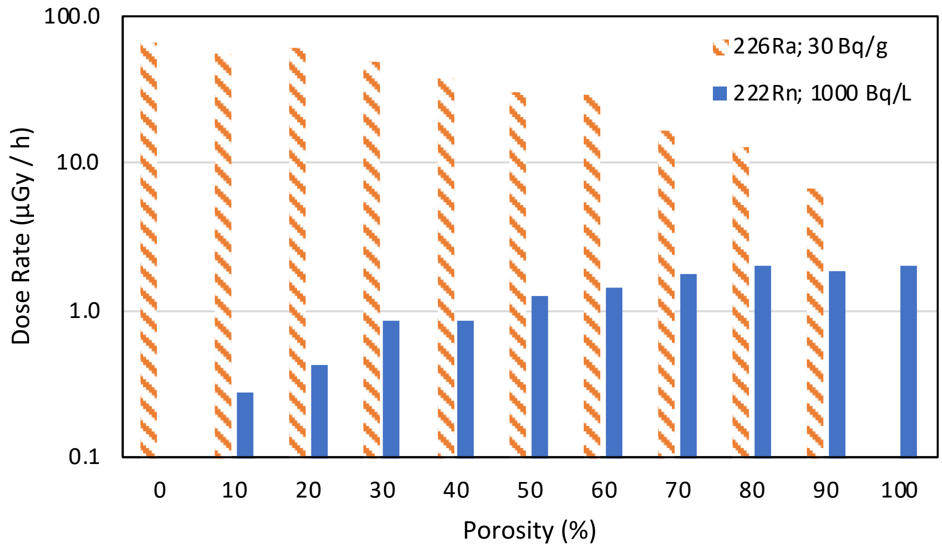

Fig. 1. *Dose rate to the diatoms as a function of porosity in the sediments*

The impact of the internal and/or external exposure to the dose rate received by the diatoms was also evaluated using GATE. Considering again the mean measured values of 1000 Bq/L ^{222}Rn activity concentration in the water and 30 Bq/g ^{226}Ra specific activity in the sediments, an increase of the dose rate is observed in the case of the combined internal/external exposure. This result can be interpreted by the fact that both ^{222}Rn and ^{226}Ra are alpha emitters of relatively small energies (5.5 MeV and 4.8 MeV respectively) which deposit their energy in a few tens of micrometers. While their energy range in the water is well known [23], in the case of the sediments the simulation offers a first evaluation of the range of the alpha particles in this specific medium.

The nanodosimetric simulations using Geant4-DNA in the case of 1000 Bq/L ^{222}Rn activity concentration in the water, showed a specific energy rate of 0.023 μGy/h for the nucleosome and 8.4 10^{-4} μGy/h for 10 DNA base pairs. The 2-orders-of-magnitude difference can be attributed to the different sizes of the modelled molecules.

For the case of the simulation of the direct DNA damage using the clustering algorithm, 131 SSBs per particle and 57 DSBs per particle were predicted. These preliminary results can only offer an order of magnitude for the DNA damages expected in the diatom.

Finally, a fisrt evaluation of the evolution of the chemical species during radiolysis of water containing ^{222}Rn was performed. In *Fig.* ***2***, the radiolytic yield (G-value, eq. 3) in a time frame of 1 μs is presented for the hydroxyl radical (OH^- **)** on the left and for the hydrogen peroxide (H_2O_2) on the right plot. The trends here represent the very early production of OH^- which is followed by a decrease as a function of time as it interacts chemically with other species, as well as, the respective production of H_2O_2.

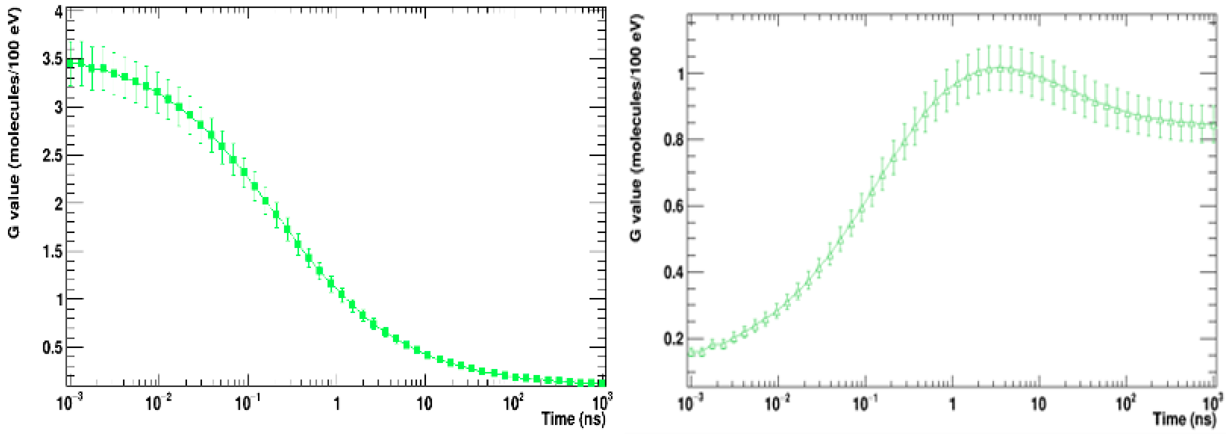

Fig. 2. *The evolution of the radiolytic yield as a function of time for $OH^{\cdot}$ (left) and H_2O_2 (right)*

CONCLUSIONS

In the current work, the effects of natural radioactivity on microorganisms living in radioactive mineral springs were simulated using GATE and GEANT4-DNA open source Monte Carlo simulation codes. This study contributes to the evaluation and the understanding of the doses received by diatoms in the mineral waters in Auvergne region in France.

In total, 27 mineral springs have been sampled and analyzed for their radiological content. Using γ-spectroscopy, the activity values of radon in the water and radium in the sediments were, among others, determined. The highest ^{222}Rn activity concentration measured in the water was 4000 Bq/L, while the highest ^{226}Ra specific activity measured in the sediments was 52 Bq/g.

Considering the measured activity values, a risk assessment for aquatic freshwater ecosystems was performed using the ERICA tool. A high risk imposed to the phytoplankton in 71% of the mineral springs was observed as a result of the high ^{226}Ra concentration in the sediments.

GATE opensource Monte Carlo platfom has been used to conduct microdosimetric simulations. Among others, the results have shown that the highest contribution to the dose received by the diatoms is due to the α-particles present in the natural decay series. It was also evaluated that the most important contributor to the dose is the ^{226}Ra which is present in the sediments of the mineral springs. The influence of the external or a combination of internal and external radiation exposure to the dose receives by the diatoms has also been studied, highlighting the necessity of further investigations.

Studies are currently performed on the capacity of the radionuclides to penetrate the silicate frustule of the diatoms.

The simulated impact of the sediment composition on the diatoms' received dose has also provoked the launch of measurements for the complete characterization of the chemical contents in the sediments.

Geant4-DNA nanodosimetric simulations have given an order of magnitude for the specific energy rate on the DNA of the diatoms. The values range between 0.023 μGy/h and 8.4 10^{-4} μGy/h as a function of the size of the modelled molecule for a ^{222}Rn source isotropically distributed in water.

In this study, the calculated SSBs and DSBs due to the presence of radon in the water are preliminary results that need to be consolidated with additional indirect damage caused by chemical species after radiolysis. The evaluation of the chemical species for a ^{222}Rn source has been performed and it needs to be supplemented with the respective evaluation for a ^{226}Ra source.

Finally, the response of the diatoms to external alpha and X-ray beams is currently being investigated in an effort to disentangle the radiation stress from other environmental stresses.

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