Review

The role of physics in radioecology and radiotoxicology

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This article gives an overview of physical concepts important for radioecology and radiotoxicology to help bridge a gap between non-physicists in these scientific disciplines and the intricate language of physics. Relying on description and only as much mathematics as necessary, we discuss concepts ranging from fundamental natural forces to applications of physical modelling in phenomenological studies. We first explain why some atomic nuclei are unstable and therefore transmute. Then we address interactions of ionising radiation with matter, which is the foundation of both radioecology and radiotoxicology. We continue with relevant naturally occurring and anthropogenic radionuclides and their properties, abundance in the environment, and toxicity for the humans and biota. Every radioecological or radiotoxicological assessment should take into account combined effects of the biological and physical half-lives of a radionuclide. We also outline the basic principles of physical modelling commonly used to study health effects of exposure to ionising radiation, as it is applicable to every source of radiation but what changes are statistical weighting factors, which depend on the type of radiation and exposed tissue. Typical exposure doses for stochastic and deterministic health effects are discussed, as well as controversies related to the linear no-threshold hypothesis at very low doses.

KEY WORDS: ecology; radionuclides; ionising radiation; toxicity

Radioactivity is everywhere around us. We cannot detect it by our senses but we have been exposed to it during our evolution no less than to other natural phenomena. This raises a number of questions on its role in the environment, which are being addressed by a scientific discipline called radioecology. In addition, exposure to nuclear radiation from natural and artificial sources implies a necessity to investigate the related health effects, and these issues are addressed by radiotoxicology. Studies of different aspects of radioactivity are no longer in the domain of physics alone, since radioecology and radiotoxicology also involve a wide spectrum of sciences ranging from chemistry to medicine. However, the concepts of nuclear quantum phenomena related to radioactivity generally still elude non-physicists. The goal of this review is therefore to help bridge this gap by summarising physical models of relevance to radioecology and radiotoxicology and help researchers from different fields to understand the microscopic phenomena that underlie macroscopic manifestations of ionising radiation.

Since the discovery of radioactivity by Henry Becquerel in 1896 (1), nuclear physics has explained a vast number of intricate interactions and processes involving nuclear forces. For the last few decades the interest has shifted towards processes which occur at energies that are much higher than those typical of an atomic nucleus. In this sense, one could say that the physics of radioactive decay is a relatively old science, but this does not hold true for radioactivity-related ecological or toxicological issues. For example, the conversion of an atomic nucleus into another, which is termed nuclear transmutation, has a twofold effect in a tissue. First, γ photons and/or charged α or β particles are emitted into a complex organic environment where they ionise atoms and molecules, change their chemical properties, and consequently the reactions they undergo. This, in turn, alters biological functions. Second, the emitting atom mutates into another one and changes its own chemical properties, which affects biological functions at the place of emission. On a larger scale, in ecosystems, propagation pathways of radionuclides through different media are of primary importance, but the exposure of humans and biota to radioactive sources in the environment brings us back to the effects of radioactivity at a molecular level. The above examples demonstrate how much the physics of radioactivity is intertwined with other sciences involved in radioecology and radiotoxicology and how this seemingly old branch of physics has now taken a new role in interdisciplinary research.

We shall first address the properties of nucleons – the proton (p) and the neutron (n) – and the forces which underlie their interactions in a nucleus. This will be followed by a discussion of the exponential law of radioactive decay, which always holds true, irrespective of the type of transmutation. We shall also give an overview of transmutations and the consequent radiation emissions. This part will be concluded by an outline of interactions between nuclear radiation and matter, which is a key to understanding the general effects of ionising radiation.

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The above considerations will be extended to radioecology, where physics provides a link between radionuclides in the environment and their sources. This takes into account not only the origin of radionuclides (primordial, decay chains, cosmogenic, anthropogenic) but also other properties which determine their distribution in the environment.

As physics also plays an important role in radiotoxicology, we shall review standard models accounting for radiation doses received via external and internal exposure. These models include the biological half-life of a radionuclide inside the body (not only its physical half-life) and the type of radiation, and also take into account that different tissues respond to exposure differently. Medical procedures (diagnostic and therapeutic) involving radionuclides and other sources of ionising radiation employ the same concepts, which represents another extension of physics into other areas of science and technology.

THE PHENOMENON OF RADIOACTIVITY

Basic concepts

There are four fundamental forces known to humankind, and they are responsible for all processes and interactions that our science has encountered (2). The first is the strong nuclear force (SNF), which underlies the attraction between nucleons and does not depend on whether the nucleons are p or n. This is the strongest force known to us, but it acts over very short distances, so that nucleons must virtually touch each other in order to bind together. Charged particles interact via the electromagnetic force (EMF) - the second of the fundamental forces - which can be either attractive (for unlike charges) or repulsive (for like charges). Since the proton carries a single positive charge, the p-p repulsion in a nucleus acts against the attractive SNF. At the same distance, the SNF is about hundred times stronger than the EMF, but the range of the latter one is infinite. Not only is the EMF important in the atomic nucleus, but is also responsible for chemical bonding that occurs at the level of valence electrons. The third of the fundamental forces is the weak nuclear force (WNF) which is responsible for beta (β) decay and, therefore, important in nuclear transmutations. It is much weaker than the SNF (10^3-10^{13}) times, estimates vary) and also short-ranged. The fourth of the fundamental forces is the gravitational force which has an infinite range but is very weak (10³⁵-10⁴² times weaker than the SNF) and therefore irrelevant in microscopic interactions.

The interplay between the SNF, EMF, and WNF determines the total energy of a nucleus and, consequently, its stability. The key factor is the number of n relative to that of p. A nucleus which has a mass number Y, comprising Z protons and Y - Z neutrons, is stable against a transmutation only if Z and Y - Z are such that they lie

within the so-called valley of stability where the ratios of n and p are balanced (3). Otherwise, the nucleus is unstable because it can transmute into a lower energy one. Unstable isotopes are termed radionuclides. For instance, ²⁰⁶Pb, ²⁰⁷Pb, and ²⁰⁸Pb are stable whereas ²¹⁰Pb, ²¹¹Pb, ²¹²Pb, and ²¹⁴Pb are not; all of them are naturally occurring, but the latter ones are radionuclides which undergo transmutations until reaching the valley of stability. There are numerous nuclear transmutations, which will be addressed later, but all of them follow a remarkably simple statistics known as the law of radioactive decay, as follows. The number *N* of radioactive nuclei is reduced in time *t* according to

$$N(t) = N_0 e^{-\lambda t},$$
[1]

where λ is the decay constant. At $t = T_{1/2} = \ln 2/\lambda$, the number of nuclei is reduced to $N_0/2$, and $T_{1/2}$ (called half-life) is traditionally used as a time scale for the decay of a radionuclide. In radioactivity studies, the most important measurable quantity is radionuclide activity A which is defined as

$$A(t) = - dN/dt = \lambda N_0 e^{-\lambda t} = A_0 e^{-\lambda t} = \lambda N(t).$$
 [2]

The SI unit for A is becquerel (Bq), and 1 Bq=1 s⁻¹. When one wants to quantify the radioactivity of a given medium that comprises both stable isotopes and radionuclides, one uses activity concentration, that is, A per unit of quantity (for instance, Bq kg⁻¹ or Bq m⁻¹).

It frequently happens that there is a cascade of transmutations instead of only one. In this case, for k members of the cascade, the time dependence of the number of nuclei is given by k coupled Bateman equations (4)

$$dN_{1}/dt = -\lambda_{1}N_{1}, dN_{i}/dt = \lambda_{i-1}N_{i-1} - \lambda_{i}N_{i}, (i = 2, ..., k).$$
 [3]

The first of the above equations describes the decay of a parent radionuclide, whereas the second accounts for the number of the nuclei of the *i*th radionuclide. $N_i(t)$ decreases due to own decay but also increases due to the decay of the previous radionuclide in the cascade. These cascades are usually termed decay chains; they are named after the parent radionuclide, and end with a stable nucleus with $\lambda_k = 0$.

The above consideration explains how to quantify a source of nuclear radiation but it neither addresses the energy carried by the radiation nor its deposition in case of an interaction with matter. The total energy $E_{\rm em}$ emitted by a radioactive source is given by

$$E_{\rm em} = t [\Sigma_{\alpha,j} A_{\alpha} \eta_{\alpha j} \varepsilon_{\alpha j} + \Sigma_{\beta,l} A_{\beta} \eta_{\beta l} \varepsilon_{\beta l} + \Sigma_{\gamma m} A_{\gamma} \eta_{\gamma m} \varepsilon_{\gamma m}].$$
 [4]

where A_{α} , A_{β} , and A_{γ} are activities resulting in emissions of α , β , and γ radiation, respectively, with indices *j*, *l*, and *m* accounting for statistically distributed emissions (with energies ε and the corresponding emission probabilities $\eta \leq 1$, which will be discussed in more detail later). For interactions between ionising radiation and matter it is important how much energy $E_{dep} \leq E_{em}$ is deposited by the radiation in a mass M, and the quantity that accounts for that is absorbed dose

$$D = dE_{\rm dep}/dM.$$
 [5]

It is also important at what rate is E_{dep} deposited, as short-term and long-term exposures to the same amount of energy may have different effects on living organisms. Therefore, often it is the dose rate

$$\dot{D} = dD/dt$$
 [6]

that is studied. The SI unit of *D* is grey (1 Gy=1 J kg⁻¹), and of \dot{D} , of course, Gy s⁻¹. However, *D* is in practice often expressed in Gy h⁻¹.

Different tissues respond differently to the same type of radiation and amount of deposited energy. In order to account for that, the concept of equivalent dose H has been introduced. H is basically D modified for specific interactions of different tissues with different types of radiation. Equations [5] and [6] apply to H in the same way as they do to D, but the unit of H is sievert (1 Sv=1 J kg⁻¹). Although Gy and Sv are dimensionally the same, they have different meanings, since Sv takes into account biological effects and Gy does not.

The above concepts apply in every situation where radionuclides undergo transmutations and emit radiation and they are in fact sufficient for understanding the meaning of the result of a measurement relevant to radioecology and radiotoxicology. However, it is also often necessary to understand the mechanisms behind such results, which requires a deeper insight into the physics of radioactive decay.

Nuclear transmutations and radiation emission

Nuclear processes are governed by the laws of quantum physics; they obey the universal conservation laws and are affected by the Einstein's energy-mass equivalence. One should therefore address these three topics before turning to nuclear transmutations and radiation.

The first step is to clarify that there are three classes of particles of importance for the issues under discussion. This first of these are photons, massless quanta of electromagnetic radiation. This class includes γ rays, whose energy ranges roughly from 10 keV to a few MeV, where 1 eV=1.6 x 10⁻¹⁹ J is the usual energy unit in microscopic processes. X-ray photons may have the same energies as γ photons, but their source is different, as they originate in electromagnetic phenomena involving electrons outside the nucleus.

The second class of the particles of interest are leptons. These particles are affected by the WNF but do not interact via the SNF. They have antiparticles of the same mass and, if a lepton is charged, of the opposite charge. The electron e⁻ and the positron e⁺ are the best know examples of a lepton and its antiparticle, respectively. They are assigned a lepton number which equals +1 for leptons, -1 for their antiparticles, and 0 for non-leptons. Conservation of lepton number is one of the six fundamental conservation laws and must be satisfied in every process. For example, the equation of β decay, i.e., of an n becoming a p, is

$$n \rightarrow p + e^{-} + \overline{v_{e}},$$
 [7]

where v_{e}^{-} is the electron antineutrino, a very light, electrically neutral lepton and the antiparticle of the electron neutrino v_{e} . The appearance of v_{e}^{-} is required because e has a lepton number +1, p and n are not leptons and their lepton numbers are therefore zero, and a lepton antiparticle is required in Eq. [7] in order that the total lepton number remains zero after the n-to-p conversion. Another conservation law explains why e and not e⁺ appears on the right-hand side of Eq. [7], and this is conservation of electrical charge: as n has no electrical charge, an opposite and exactly equal charge must cancel the positive charge of p.

Finally, we ought to discuss baryons, i.e., particles that consist of quarks held together by the SNF. They are heavy particles, can be either electrically charged or neutral, have antiparticles, and, like leptons, are assigned a baryon number (+1 for particles, -1 for antiparticles, 0 for nonbaryons). The proton and the neutron belong to this class, both being composed of three quarks. There is also the law of conservation of baryon number, which is satisfied in Eq. [7], because both n and p have a baryon number +1 and the equation is balanced with regard to baryon number.

 β^+ decay, which is a p-to-n conversion, is a good example of the energy-mass equivalence that is often manifested in nuclear processes. An isolated p cannot become an isolated n because of its slightly smaller mass; thus, β^+ decay seems to be impossible. However, there are β^+ decays in nature. For instance, ²³Mg transmutes to ²³Na, which is accompanied by the emission of an e^+ and a v_e (which means that the lepton number, baryon number, and electrical charge are conserved). The process which allows for that to happen is the conversion of some of the nuclear mass in ²³Na into the nuclear binding energy according to $E = Mc^2$, where c is the speed of light. This apparently makes the resulting neutron lighter and enables the p-to-n conversion. In turn, the classical law of conservation of mass is actually just a manifestation of the fundamental law of conservation of energy.

The remaining topic that we ought to address is the quantum nature of the atomic nucleus. It turns out that energy levels in atomic nuclei are quantised, i.e., discrete. This is because the manifestations of the SNF are always quantum. What is important for this discussion is that a nucleus can be in an excited energy state and fall into a lower state not only by transmutation but also by emitting a γ photon without changing either the total number or the

type of its nucleons. This is similar to the emission of a photon when an electron in an atomic shell falls into a lower energy state, so that the total energy of the atom is reduced by this emission. In the case of nuclei, this often happens (but not always) after a transmutation has taken place, if the daughter nucleus is in an excited state and then relaxes until it reaches the lowest energy state by γ emission(s). Therefore, a transmutation can occur with or without an emission of γ radiation, depending on the nucleus.

While details of the outlined phenomena are generally complex, the above simplified discussion is sufficient to understand different types of nuclear transmutations and consequent emissions of radiation, at least qualitatively. It should also be noted that v_e and $\overline{v_e}$ are of no interest regarding the ecological and toxicological effects of nuclear radiation, as they react with matter only weakly, by the short-ranged WNF. We shall therefore focus on the emission of γ photons and α , β -, and β + charged particles.

Transmutations that result in the emission of an α particle (which are the same as the ⁴He nuclei, consisting od two p and two n and having a double positive charge) occur mainly for heavy nuclei (Y > 150) rich in protons (5). Energies of α particles are high (several MeV), discrete, and characteristic of a given nucleus (5, 6). Therefore, the emitting nucleus can be identified and its activity quantified by means of α -ray spectrometry due to its unique α -particle energy signature. The emission of α particles by an ensemble of the same nuclei may be statistically distributed. For example, in the α decay of ²³⁸U the emitted α particles have $\varepsilon_{\alpha 1} = 4.198$ MeV ($\eta_{\alpha 1} = 0.77$) and $\varepsilon_{\alpha 2} = 4.151$ MeV ($\eta_{\alpha 2} = 0.23$), see Eq. [4] (6).

In β decay, n transforms into p and vice versa, depending on the position of a radionuclide relative to the valley of stability (3). There is a good reason for using the names β^+ and β^{-} particles for e⁺ and e⁻ created in β decay, since this clarifies their origin and explains their high energy (which is still generally lower than that of α particles). The spectrum of β radiation is, in contrast to that of α radiation, continuous in energy, since some of the energy is taken by v_{e} or v_{e} . A β -emitting radionuclide is usually characterised by the average and maximum energies of the emitted β particles. the former being roughly 1/3 of the latter (7). Therefore, one must take into account this in Eq. [4] with regard to ε_{β} . The statistical branching of emission energies is present in β decay as well. For instance, ¹³⁷Cs emits two types of β particles when transmuting, with the average energies of 156.8 keV ($\eta_{B1} = 0.946$) and 415.2 keV ($\eta_{B2} = 0.054$). Some radionuclides undergo both α and β decay; for instance, naturally occurring ²¹²Bi exhibits an α decay into ²⁰⁸Tl in 36% of the cases, and a β decay into ²¹²Po in 64%.

Due to the quantisation of nuclear energy levels, γ photons have well defined energies, and this is the basis of γ -ray spectrometry, which is the most potent experimental method in radioecology, able to identify radionuclides and quantify their *A* more efficiently than α -ray spectrometry. The number and statistical distribution of γ emissions vary

from nucleus to nucleus. For instance, there is no γ emission that accompanies the transmutation of ⁹⁰Sr into ⁹⁰Y, whereas when ¹³⁷Cs transmutes to metastable ¹³⁷Ba, there are four gamma emissions with 31.82 keV $\leq \varepsilon_{\gamma m} \leq 661.62$ keV and $0.0022 \leq \eta_{\gamma m} \leq 0.8462$. There are also very complex situations, such as those in the decay of ²¹⁴Bi, where the number of emissions is close to two hundred, with 76.86 keV $\leq \varepsilon_{\gamma m} \leq 3269.7$ keV and 9.8 x 10⁻⁵ $\leq \eta_{\gamma m} \leq 0.4609$.

Besides "classical" α and β decays, there are also other nuclear processes that result in radiation emission. A transmutation similar to β^+ decay is electron capture, where a p in a nucleus captures an e from the atomic shell, transforms into an n and emits a v_{a} , which may also result in the emission of γ photons until the lowest energy level of the nucleus is reached. In some processes, p or n are emitted from a nucleus, and in spontaneous or induced fission, smaller nuclei (fission fragments) are emitted together with p and n. There are also radiation emissions that change neither Y nor Z. These are termed internal conversion and isomeric transition. Isomeric transition is similar to the regular emission of a γ photon. The only difference is in the time scale involved: while a γ photon is normally emitted due to the relaxation of a nucleus from an excited state that lasts about one picosecond, in isomeric transition the metastable state lasts longer than a nanosecond. In internal conversion, the relaxation of a nucleus from an excited state does not cause γ photon emission, but an ejection of an e-from the atomic shell.

Nuclear processes which result in the emission of radiation are summarised in Table 1. They are known from nuclear physics, whereas radioecology and radiotoxicity

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|---------|-------|------------|-----------|----------|
| Table 1 | Modes | of nuclear | radiation | emission |
| | | | | |

| Mode | Emitted radiation | Daughter nucleus |
|--------------------------|--|-------------------------|
| α decay | α | Y - 4, Z - 2 |
| p emission | р | Y - 1, Z - 1 |
| n emission | n | Y - 1, Z |
| double p emission | 2p | Y - 2, Z - 2 |
| spontaneous fission | smaller nuclei and nucleons | |
| induced fission | smaller nuclei and nucleons | |
| β ⁻ decay | e^{-} and v_{e}^{-} | <i>Y</i> , <i>Z</i> + 1 |
| β^+ decay | e^+ and v_e | <i>Y</i> , <i>Z</i> – 1 |
| electron capture | V _e | <i>Y</i> , <i>Z</i> – 1 |
| double β^{-} decay | 2e ⁻ and 2 $\overline{v_e}$ | <i>Y</i> , <i>Z</i> + 2 |
| double β^+ decay | $2e^+$ and $2v_e$ | Y, Z - 2 |
| double electron capture | 2 <i>v</i> _e | <i>Y</i> , <i>Z</i> – 2 |
| isomeric transition | γ | <i>Y</i> , <i>Z</i> |
| internal conversion | e ⁻ from atomic shell | <i>Y</i> , <i>Z</i> |

Y and Z refer to the mass number and charge of the mother nucleus. All of the listed modes may result in an additional emission of γ photons

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address the impact of this radiation on the environment and organic matter.

Interactions of nuclear radiation and matter

Several types of highly energetic radiation may be emitted due to nuclear processes, which may result in the ionisation of atoms and molecules, if matter is exposed to such radiation. The most abundant products of radioactive decay are α , β , and γ rays, and they interact with matter differently. The effects of α and β particles are similar, because they are charged and interact with electrical charge along their paths via the long-range EMF. As they are passing through matter, they lose energy, as described by the Bethe formula, which states that the loss of energy per travelled distance is proportional to the square of the charge of a particle (8). This energy is transferred to atoms and molecules of the exposed material, causing ionisation. However, the charge of α particles is +2, whereas that of β particles is -1 or +1, which means that α particles interact with matter stronger and lose energy faster than β particles. Because α particles are heavy, their trajectories are straight and their stopping distance in a given material is a function of energy only. By contrast, β particles change direction as they collide with atoms and molecules, which results in a wiggly path and a variable stopping length. Alpha particles deposit more energy into matter than β particles because of their larger energies, and their stopping distance is shorter. The energy of α particles is between 4 and 9 MeV, and in water they stop after 30–150 μ m. The same intervals for β particles are typically 0.15-3.5 MeV and 0.03-1.7 cm, respectively (9).

Since γ rays consist of photons, that is, of massless quanta of electromagnetic radiation, they interact with matter differently, via three mechanisms. The first is Compton scattering, i.e., the inelastic scattering of a highenergy photon on an electron. This results in the ionisation of an atom or a molecule, while the photon loses energy but continues to propagate after the scattering. The second mechanism is photoelectric effect, in which a photon is completely absorbed, and this also causes ionisation. The third mechanism is the production of $e^{-}e^{+}$ pairs in the vicinity of a heavy nucleus. In this process, a photon with a minimum energy of 1.022 MeV is converted into an e-e+ pair. Since antimatter is not stable, the e⁺ soon annihilates with an e-, which results in the creation of two photons with an energy of 511 keV each. They have exactly this energy, because it corresponds to the mass of e⁻ and e⁺, which is required by the mass-energy equivalence and the law of conservation of energy. These photons can afterwards cause more ionisations. Generally, the penetration of γ radiation into matter is much stronger than that of α and β radiation.

Interactions of v_e and $\overline{v_e}$ with matter are marginal, but this is not the case with protons and neutrons which may also be emitted in nuclear processes. Just like α and β particles, protons interact with matter mainly due to its charge and the long-ranged EMF. Their interactions with nuclei via the short-ranged SNF are much less frequent. Therefore, protons behave in a way similar to α and β particles. Neutrons are, however, electrically neutral and can therefore interact only via the SNF or the magnetic component of the EMF (which is a weak interaction that cannot cause ionisation). By consequence, neutrons travel long distances through matter. However, when they interact with a tissue, the health consequences may be significant.

RADIONUCLIDES IN THE ENVIRONMENT

Radioecology is a scientific discipline which addresses the presence and propagation of radionuclides in natural and agricultural systems populated by humans, animals, and plants. It studies the impact of environmental radioactivity on living organisms at the level of populations and communities and also uses radionuclides as markers in studies of different ecological processes (10). Radioecology rests on interdisciplinary research of the distribution and ecological impact of radionuclides in the environment. The distribution of a radionuclide in the environment is determined by its chemical properties which are set by valence electrons. Its ecological impact depends on the radiation emission, which is a nuclear property.

Naturally occurring radionuclides in the environment

Numerous radionuclides have been identified so far, of which at least 3000 have been characterised experimentally (3). However, a large percentage of them are of no interest to radioecology, since they either decay rapidly or are not present in the environment in considerable quantities (unless there has been a leakage from a facility where they have been produced, used, or stored). Generally, radionuclides can be naturally occurring or anthropogenic.

There are numerous naturally occurring radionuclides, and many of them originate from primordial radionuclides which have been formed in massive stars. All primordial radionuclides have very long half-lives, the shortest being 704 million years of 235 U and the longest 2.41 x 10^{24} years of ¹²⁸Te. Four of them are very important in radioecology: 40 K ($T_{1/2}$ =1.25 x 10⁹ years), 235 U, 238 U ($T_{1/2}$ =4.47 x 10⁹ years), and ²³²Th ($T_{1/2}$ =1.40 x 10¹⁰ years). Of these, ⁴⁰K is of special importance for biological systems, as it accounts for 0.012% of all potassium, which is abundant in all living organisms. This means that the activity of 1 g of K is about 31 Bq (11), and it is a constant internal source of radioactivity in humans and biota. 235U and 238U also have fixed isotopic abundances, 0.72% and 99.275%, respectively. ²³²Th, ²³⁸U, and ²³⁵U are parent radionuclides of their respective decay chains. These chains comprise 12-15 radionuclides and end with stable isotopes of lead. Elevated concentrations of the chain members in a tissue can be harmful not only because of ionising radiation emission but also because of the chemical toxicity of heavy metals. In addition, the chains comprise some radium isotopes that have a twofold negative effect on living organisms. They emit α particles, which makes them rather dangerous in internal exposure, and they transmute into isotopes of radon, a noble radioactive gas that can easily enter a body by inhalation.

A special class of naturally occurring radionuclides are cosmogenic radionuclides. Most of them are produced in the atmosphere, in collisions of cosmic rays with the nuclei of atmospheric gasses (12), and some even in the lithosphere (13), which is termed cosmogenic nucleosynthesis. They are relatively light elements with short to moderate halflives, and they descend towards Earth as part of aerosols and/or precipitation. For organic matter the most important is ¹⁴C which, like ⁴⁰K, acts as an omnipresent source of radioactivity in living organisms. Other radionuclides of that kind can be used to study solar activity, which affects the flux of cosmic rays and consequently cosmogenic nucleosynthesis. These radionuclides are γ emitters such as ⁷Be and β emitters such as ³⁹Ar, ³²P, ³³P, ³⁵S, and ¹⁰Be (14–17). Although cosmic rays consist of very energetic particles, their flux at the Earth's surface is very weak, and this radiation poses no threat for living organisms.

Organisms on Earth have evolved in a moderately radioactive environment and have developed mechanisms to cope with radioactivity up to a certain dose. However, technological processes may result in enhanced concentrations of naturally occurring radionuclides, which then may become as dangerous as any other source of ionising radiation. This may happen in industrial processes where materials from nature, always containing a certain amount of radionuclides, are used. Good examples are oil industry (18) and the production of soil amendments for use in modern agriculture (19), but these industries are regularly monitored for radioactivity. In other words, naturally occurring radionuclides pose no threat unless their concentrations increase to abnormal levels over long periods of time. The main danger comes from anthropogenic radionuclides, which are normally not present in the environment.

Anthropogenic radionuclides in the environment

The most important source of anthropogenic radionuclides is the fission of uranium in power plants, nuclear-powered ships, and nuclear weapons tests. Some, on the other hand, are produced in controlled reactions under laboratory conditions, for instance in particle accelerators. The fission of uranium results in numerous radionuclides. Some are heavy and long-lived, such as 239 Pu, 240 Pu, 241 Pu, and 241 Am. The *Y* of most of the others is between 80 and 100 or between 130 and 150, with their half-lives ranging from a few days to several tens of years.

The threat due to the presence of a radionuclide in the environment is determined by its abundance, role in living organisms, and pathways of propagation through different media. By these criteria, the most dangerous of anthropogenic radionuclides are the uranium fission products ⁹⁰Sr (β emitter, $T_{1/2}$ =28.9 years), ¹³¹I (β and γ emitter, $T_{1/2}$ =8.03 days), ¹³⁴Cs (β and γ emitter, $T_{1/2}$ =2.06 years, and ¹³⁷Cs (β and γ emitter, $T_{1/2}$ =30.2 years). They are all produced in large quantities, can enter living organisms easily by ingestion and inhalation, and can mimic some biogenic elements. 90Sr can substitute calcium, 131I non-radioactive iodine, and ¹³⁴Cs and ¹³⁷Cs potassium. In addition, the presence of ¹³⁴Cs is a clear indicator of a nuclear accident, since this radionuclide is not produced in nuclear explosions. Although the half-life of ¹³¹I is short, it poses a threat because it does not substitute another atom (like Sr substitutes Ca) but directly takes the role of non-radioactive iodine. Moreover, carried by air masses, it can travel long distances before decaying [for instance, its presence was measured in Zagreb, Croatia, after the Fukushima accident (14)]. Both 90Sr and 137Cs have long half-lives, and this is the reason why they are of special interest in radioecological studies. Although the main releases of ¹³⁷Cs and ⁹⁰Sr occurred relatively long time ago, during nuclear weapon tests and the Chernobyl disaster, these dangerous radionuclides are still abundant in the upper atmosphere from whence they descend towards the surface and enter the ground-level environment.

Besides their generally negative ecological impact, anthropogenic radionuclides may also have a positive role. They can be used in different technological processes, as well as in medical diagnostic and therapeutic procedures. For example, ⁶⁰Co and ¹³⁷Cs are used in gamma-ray radiography for cargo scanning. ¹³¹I is widely used in medical procedures related to thyroid gland. ⁶⁰Co, ¹³⁷Cs, ¹⁹⁸Au, ¹⁹²Ir, ¹²⁵I, and ¹⁰³Pd are used in brachytherapy (20). ⁶⁰Co is usually the source of radiation for the gamma knife. ^{99m}Tc, ¹²³I, ¹¹¹In, ²⁰¹Tl, and ⁶⁷Ga are used in single-photon emission tomography, while positron emission tomography is based on the use of ¹⁸F, ¹¹C, ¹⁵O, ¹³N, and ⁶⁴Cu (21). Medical and technological procedures employing radioactive sources continue to develop.

Once used, radioactive matter must be disposed in a safe and ecologically harmless way, and there is much concern about that. This is, however, a social and legal problem, not a technological one. The long-term disposal of fission products has been technologically solved, and the major threat lies in misuses of regulations and standards, which may range from simple negligence to criminal activities leading to unauthorised dissemination of radioactive matter. Radiopharmaceuticals must be very active and they therefore decay rapidly, so there are no problems with their long-term storage. They might become a threat only if they are not safely transported from production facilities to medical institutions or if the safety procedures during application are not implemented to the letter. In other words, we have developed technology for the safe handling of radioactive matter, and radioecological concerns should address mishandling and misuse of the existing regulations and procedures.

EFFECTS OF IONISING RADIATION ON LIVING ORGANISMS

Qualitative considerations

According to a 1963 definition by the International Atomic Energy Agency (IAEA), the toxicity of a radionuclide is "...the ability of the nuclide to produce injury, by virtue of its emitted radiation, when incorporated in a body" (22). This early definition of radiotoxicity took into account only the hazard due to a radionuclide being inside a body. Nowadays, however, the study of damage caused by ionising radiation is no longer limited to internal sources, but includes external sources such as nuclear radiation, X-rays, and even UV radiation, all of which have lately been on the rise. It therefore seems more appropriate to use the terms radiotoxicity and radiotoxicology in a wider sense that encompasses internal and external sources and every radiation that may cause ionisation in organic matter. However, one still cannot regard radiation from a source inside a body on par with radiation coming from the outside. Skin, for example, is effective in stopping α and β radiation, and external irradiation by α and β particles over skin is considered less threatening, even though β particles may cause radiation burns. By contrast, α particles from internal sources are extremely dangerous, as there is no skin to stop their strong interactions with tissue and the consequent deposition of large amounts of energy.

Ionising radiation has enough energy to break chemical bonds and alter chemical properties of atoms and molecules, which in living organisms leads to changes in local biological mechanisms. The local deposition of energy is the main cause of irreparable tissue damage, whereas the same energy absorbed over a larger volume causes milder effects that can be repaired. This also sheds more light on the fact that α particles cause more damage than β or γ radiation, since their stopping distance is short and the deposition of energy very local. In contrast, β particles and γ (or X) photons scatter several times during their travel through a tissue and cause ionisation over a larger volume. The time of exposure has a similar effect: the absorption of a given radiation dose is more dangerous if the exposure time is short, since biological repair mechanisms are more effective over long time. For instance, to absorb about 10 Sv over a few hours can be lethal, whereas this equivalent dose absorbed over a lifetime is usually harmless.

In order to induce noticeable effects in a living organisms, ionisation by high-energy particles and photons must alter the functioning of a large number of cells and/ or their ability to divide. This, of course, involves complicated processes, but two of these can be singled out for their dominant effects. The first is direct ionisation of DNA, and the second is ionisation of water molecules (which are the most abundant in a cell) and other organic molecules (23). The latter mechanism affects the DNA indirectly as well, and this is why the overall consequences of exposure to ionising radiation are mainly related to DNA damages (23, 24). These damages can be reparable and irreparable, may cause mutations and chromosomal aberrations, lead to cell death or mitotic inhibition, etc. Of course, this may have severe consequences, such as radiation sickness or cancer, but also helps in oncologic radiotherapy and related procedures. Cells which divide faster are more sensitive to the described processes, and this means that different tissues respond to ionising radiation differently.

Radiotoxicological relevance of a radionuclide depends on its abundance and $T_{1/2}$, chemical properties that determine the pathways of its propagation through the environment and uptake by living organisms, type and energy of the radiation it emits, possible chemical toxicity alongside radiotoxicity, and, finally, on its biological half-life T_{bio} in a human body (25). The effects of $T_{1/2}$ and T_{bio} are combined into an effective time T_{eff} according to

$$1/T_{\rm eff} = 1/T_{\rm bio} + 1/T_{1/2},$$
 [8]

which is characteristic of a given radionuclide not only because of $T_{1/2}$ but also because T_{bio} depends on its chemical and metabolic properties. The values of $T_{1/2}$, T_{bio} , and T_{eff} of selected radionuclides are listed in Table 2.

We shall first address naturally occurring radionuclides. Since ⁴⁰K is a biogenic radionuclide we have evolved with, and since its presence in the body is controlled by physiological processes, it is not considered to be a threat in internal exposure. However, its concentration may be elevated in certain industrial products, e.g., in fertilisers (19), which increases the risk of external exposure to its γ radiation. The same holds for all γ emitters within the ²³⁸U, ²³⁵U, and ²³²Th decay chains. Some members of these chains pose more threat than others. These are, first of all, isotopes of Ra and Rn, most notably those with the longest $T_{1/2}$, that is, ²²⁶Ra ($T_{1/2}$ =1600 years) and ²²²Rn ($T_{1/2}$ =3.8 days) from the ²³⁸U decay chain. ²²⁶Ra is chemically similar to Ca and can substitute it both in geological formations, including soil (26), and in organic matter. Therefore, its migrations through the environment more or less follow those of Ca,

Table 2 Characteristic times $T_{1/2}$, T_{bio} , and T_{eff} of selected radionuclides, expressed in years (y) and days (d) (25)

| Radionuclide | T _{1/2} | T _{bio} | T _{eff} |
|------------------------------------|------------------------|------------------|------------------|
| | | | |
| ³ H | 12.3 y | 12 d | 12 d |
| ¹³¹ I | 8.3 d | 138 d | 7.6 d |
| ⁹⁰ Sr | 289 у | 50 y | 18.3 y |
| ¹³⁷ Cs | 30.08 y | 70 d | 70 d |
| ²³⁸ U/ ²³⁵ U | 4.47×10 ⁹ y | 15 d | 15 d |
| ²²⁶ Ra | 1600 y | 44 y | 42.8 y |
| ²¹⁰ Pb | 22.2 y | 10 y | 6.9 y |
| ²¹⁰ Po | 138.4 y | 60 d | 41.8 d |
| ²³⁹ Pu | 24110 y | 200 y | 198.4 y |

which also applies to its uptake into living organisms. It tends to concentrate in the bones if ingested, has a long $T_{\rm eff}$, transmutes through α decay (emitting photons in the process) directly into ²²²Rn, a noble gas and an α emitter that also can be inhaled. In fact, ²²²Rn inhalation is considered to be the most serious radiological threat from naturally occurring radioactivity, with proven links to lung cancer (27). A whole cascade of radionuclides and their decays (emitting α , β , and γ radiation) follows after the decay of ²²²Rn. This eventually results in radioactive ²¹⁰Pb with $T_{\rm eff}$ =6.9 years and, finally, stable ²⁰⁶Pb, which adds the chemical toxicity of lead to its radiotoxicity due to ²²²Rn inhalation. Because of that, serious measures have been taken over the last decades to reduce inhalation exposure to ²²²Rn (28).

Of anthropogenic radionuclides, the most dangerous are fission products, as outlined before. Some of them are extremely toxic and can therefore be considered poisons, e.g., ²³⁹Pu, which is an α emitter with $T_{\rm eff}$ =198.4 years and which concentrates in the bones. However, heavy long-lived fission products entering a living organisms have quite limited pathways, and the majority of long-term health problems due to the radiotoxicity of anthropogenic radionuclides is caused by lighter fission products ¹³¹I, ¹³⁴Cs, ¹³⁷Cs, and ⁹⁰Sr (see above). Judging by the long $T_{\rm eff}$ =18.3 years (Table 2), ⁹⁰Sr is especially dangerous although it emits only β radiation, unlike the other three radionuclides which are both β and γ emitters.

Quantification through physical modelling

Physical approach to the complex effects of ionising radiation on biological systems relies on establishing a proper relation between *D* and *H*, taking into account that, in general, different tissues interact with high-energy particles and photons differently. Exposure of an organ Ω to radiation Λ (which can be α , β , γ , X, n, etc.) results in the absorption of a dose $D_{\Omega\Lambda}$. If the organ is exposed to several types of radiation, it receives an equivalent dose

$$H_{\Omega} = \Sigma_{\Lambda} W_{\Lambda} D_{\Omega \Lambda}, \qquad [9]$$

where w_{Λ} is the radiation weighting factor (29) (see Table 3). While $D_{\Omega\Lambda}$ is measured in Gy, the use of w_{Λ} allows us to introduce Sv as the unit for H_{Ω} . The total equivalent dose for the whole organism is calculated as follows:

$$H = \Sigma_{\Omega} w_{\Omega} H_{\Omega}, \qquad [10]$$

where w_{Ω} is the weighting factor accounting for the contribution of an organ Ω (29) (see Table 4) and satisfying $\Sigma_{\Omega} w_{\Omega} = 1$. The unit of *H* is also Sv.

The above considerations, supplemented with the data in Tables 3 and 4, assume that a body and/or a given organ is exposed to ionising radiation but do not specify whether exposure is internal or external. For some weakly penetrating types of radiation, such as α , β , or p, it is implicitly assumed that the source is internal. For strongly penetrating radiation, such as γ or X photons, doses from external sources received through skin are also important, and this leads to some widely used practical quantities. The first is ambient dose equivalent $H^*(10)$, representing H at 10 mm below the skin for a person occupying a given volume of space. The second is personal dose equivalent $H_{\rm p}(10)$, which is calculated for a specific person taking into account the type of radiation, geometrical aspects of exposure, etc. These quantities are important because external exposure to high-energy photons is nowadays widespread. An example is the exposure of patients and medical personnel to X and γ rays in the apeutic and diagnostic medical procedures. It has become common in medicine, industry, or cargo scanning, etc., to calibrate dosimeters to $H^*(10)$ rather than to D.

The IAEA publishes updates on numerous aspects of exposure to ionising radiation (30), and these comprehensive data are then modelled to assess the related health hazard. While modelling may change from case to case, the general approach is basically the same as that outlined above.

Table 3 Radiation weighting factors w_{Λ} as functions of energy and radiation type Λ

| Λ | Er | nergy | w_{Λ} |
|--|-----------------------|---------------------------|------------------------|
| γ | all | | 1 |
| | all | | 1 |
| β-, β+ | <1 | 5 | |
| | 10-1 | 10 | |
| | 100 ke | V–2 MeV | 20 |
| n | 2-2 | 10 | |
| | >20 MeV | | 5 |
| p | all | | 2 |
| α, fission fragments, heavy nuclei | | all | 20 |
| Fable 4 Radiation we | ighting fac | tors w_{Ω} for dif | ferent organs Ω |
| Ω | | w _Ω | Σw_{Ω} |
| bone marrow, large intestine, lungs, stomach, breasts | | 0.12 | 0.6 |
| gonads | | 0.08 | 0.08 |
| bladder, esophagus, liver, thyroid | | 0.04 | 0.16 |
| bone surface, brain, salivary gland, skin | | 0.01 | 0.04 |
| adrenal gland, thora | acic | | |
| vertebrae, gallbladd heart, kidneys, lym nodes, muscles, ora mucosa, pancreas, j small intestine, sple thymus, uterus | ph il prostate, | | 0.12 |

Radiation doses, deterministic and stochastic effects, linear no-threshold model

Life on Earth has evolved in a moderately radioactive environment. This suggests that low doses of ionising radiation do not pose a major health hazard. However, high doses may result in stochastic or deterministic effects on human health. Stochastic effects refer to health problems that occur by chance, because exposure to ionising radiation entails a higher probability for the development of a disease. A good example is cancer induction. Deterministic effects are those that are related to exposure beyond any doubt. Generally, there is a threshold dose for the deterministic occurrence of a disease; for instance, skin burns or bone marrow depression occur at an acute exposure to about 1 Sv (31). These effects also depend on the dynamics of exposure: receiving a dose over a short time, for instance during therapy, differs from prolonged exposure to an elevated level of radioactivity. In acute whole-body exposure to γ radiation in excess of 2 Sv the deterministic effect is radiation sickness. If exposure is over 20 Sv the deterministic effect is death (31).

Since it is impossible to completely avoid exposure to ionising radiation, the IAEA provides reference levels for cumulative doses that should not be exceeded. According to these recommendations, the annual dose received through public exposure should not exceed 1 mSv. For occupational or therapeutic exposure this limit is 1 to 20 mSv, and for public exposure in emergency situations 20 to 100 mSv (32).

There is a finer division of these limits, but there is also an intriguing, more general question: is there a dose threshold below which exposure to ionising radiation can be considered harmless or even beneficial for human health? This issue has been controversial for several decades already, and it underlies an ongoing debate on the nature of stochastic effects. It is widely accepted that adverse health effects of ionising radiation correlate linearly to exposure dose (33), as shown in Figure 1, but this has been evidenced only for doses exceeding a few tens of mSv (regimes II and III in Figure 1). At relatively high doses, above 100–200 mSv, adverse health effects have been well documented (regime III) whereas for lower doses, (still above 10-50 mSv; regime II), the level of confidence is not as high as for regime III. The most intriguing case is that of very low doses (regime I), especially because it is relevant to public and occupational exposure.

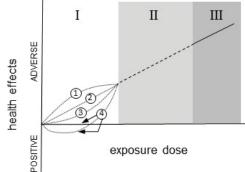
The working hypothesis of the IAEA is that any nonzero dose implies a non-zero risk of stochastic effects (30). This is called the linear no-threshold model (LNTM), and in Figure 1 it is marked by the dotted curve 2 in region I. While the IAEA generally supports this hypothesis, it is still reluctant to claim that it has been proven. In the scientific community, arguments have been presented both in favour (33) and against (34) the LNTM. Curves 1, 3, and 4 in Figure 1 show other possibilities for low exposure doses, and imply that the high-dose linear behaviour does not apply for low doses, because the adverse health effects could be either stronger (curve 1) or weaker (curve 3) than predicted by the LNTM. The most intriguing is curve 4, where health effects might either be absent or even positive, as indicated by the arrows. It is likely that the interest in exposure to very low doses will continue, and answering the outlined questions will require not only medical studies but also precise measurements and modelling of doses – which is again in the domain of physics.

CONCLUSIONS

Radioecology and radiotoxicology are scientific disciplines that tackle the complex impacts of ionising radiation on the environment, human health, and society. Radioecological studies are mainly dedicated to identifying and quantifying radionuclides in the environment as well as to determining their sources and pathways of propagation. This information is then used to evaluate the threats coming from environmental radioactivity in a given area. Radiotoxicology is focused on identifying and assessing the effects of ionising radiation on humans and biota irrespective of the source, which can be either naturally occurring or anthropogenic. It is obvious that the two disciplines must involve scientists from different fields, that is, from various natural, technical, biotechnical, and medical sciences. However, the common basis of radioecology and radiotoxicology is physics, since most processes and interactions related to ionising radiation are in its domain.

Among all sciences, physics is specific by its methodology of using a complex mathematical language to set up models that are used to describe natural phenomena and quantify their manifestations and consequences, and it

Figure 1 Health effects of ionising radiation as a function of exposure dose. The dose that corresponds to the border between regimes I and II is 10-50 mSv, and that between regimes II and III is 100-200 mSv. In regime III, adverse health effects have been well documented, while in regime II, they have been mainly identified. There is no consensus on the effects in regime I, and any of the cases 1–4 is possible. In case 1, low doses are even more harmful than the extrapolated high-dose effects represented by linear no-threshold model (case 2), whereas the opposite is argued for cases 3 and 4. Case 4 predicts positive or no effects below a certain threshold exposure dose, as indicated by the arrows



is generally difficult for non-physicists to understand physical concepts. However, the laws of physics can be brought closer to a wider audience if presented in a way that is scientifically correct but does not go into unnecessary detail. In this review, we have followed this approach and summarised physical phenomena relevant to researchers in radioecology and radiotoxicology without in-depth discussions and complex mathematics, as we believe that non-physicists who are active in radioecology and radiotoxicology may benefit from this simplification of a generally complex matter.

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Uloga fizike u radioekologiji i radiotoksikologiji

U radu je dan pregled fizikalnih koncepata važnih za radioekologiju i radiotoksikologiju radi premošćivanja procjepa koji postoji između nefizičara u tim znanstvenim disciplinama i općenito kompliciranoga jezika fizike. Koristeći se deskriptivnim pristupom, uz samo onoliko matematike koliko je nužno, prezentirana je relevantna fizika, od fundamentalnih prirodnih sila do primjena fizikalnoga modeliranja u fenomenološkim studijama. Najprije je objašnjeno zašto su neke atomske jezgre nestabilne pa stoga transmutiraju, te koje su sile, čestice i zakoni očuvanja pritom uključeni. Potom se adresiraju interakcije ionizirajućega zračenja s materijom, što je temelj i radioekologije i radiotoksikologije. Prezentirani su relevantni prirodno pojavni i antropogeni radionuklidi te su razmatrana njihova svojstva u vezi s njihovom pojavnošću u okolišu te s toksičnošću za ljude i biotu. Pritom postoje kombinirani učinci fizikalnih i bioloških poluvremena života radionuklida, što valja uzeti u obzir pri svakoj radioekološkoj ili radiotoksikološkoj procjeni. Također je dan pregled osnova fizikalnoga modeliranja koje se uobičajeno koriste u studijama zdravstvenih učinaka izloženosti ionizirajućemu zračenju, što je primjenjivo na sve izvore zračenja, ali uz korištenje statističkih težinskih faktora koji ovise o vrsti zračenja i izloženom tkivu. Razmatrane su i tipične doze izloženosti za stohastičke i determinističke zdravstvene učinke, kao i kontraverze vezane uz hipotezu o linearnom odzivu bez praga pri vrlo niskim dozama.

KLJUČNE RIJEČI: ekologija; ionizirajuće zračenje; radionuklidi; toksičnost