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TECHNOLOGICALLY ENHANCED NATURAL RADIATION

A.S. Paschoa and F. Steinhäusler

VOLUME SEVENTEEN

RADIOACTIVITY IN THE ENVIRONMENT

TECHNOLOGICALLY ENHANCED NATURAL RADIATION

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RADIOACTIVITY IN THE ENVIRONMENT

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Volume 17: Technologically Enhanced Natural Radiation
(A.S. Paschoa and F. Steinhäusler, Authors)

PREFACE

When we decided back in January 2008 to jointly write a book on Technologically Enhanced Natural Radiation (TENR) we had no clear picture of the effort that would be involved in the project. Less than one month later, Murdoch Baxter, editor of the Elsevier's Radioactivity in the Environment Series sent Anselmo a stimulating message that convinced us of the importance of writing such a book. At that time, Anselmo was actively editing the Proceedings of the 8th International Symposium on the Natural Radiation Environment (NRE-VIII), which had been held in Búzios, Rio de Janeiro, Brazil, October 7–12, 2007; as a consequence there were superimposed time intervals in which he was finishing a project and starting a new one. The initial plan for the book was modified while writing it to avoid duplication with other books, which addressed the wide field of natural radiation. The book starts with a quasi-philosophical question of whether TENR is a Universal issue or not. The answer to this question is an unequivocal *yes*. TENR as a global issue is presented as a statement rather than a question. The evolution of global exposure rate estimates since 1959 is presented to show that such estimates have grown by a factor of two. The book explores current issues like depleted uranium (DU). The World Health Organization's (WHO's) views on the DU problem are presented, and the recommendations of the Institute of Medicine (IOM) of the National Academies (of the United States) on this subject are summarized. The longest chapter of the book deals with the intricacies of terrestrial, atmospheric, and aquatic natural radioactivity. Selected industries and their relationship to naturally occurring radioactive materials (NORM), technologically enhanced NORM (TENORM), and TENR raw material product, by-product, or residues are examined with regard to their potential to enhance radiation exposure to workers, consumers, and members of the general public. The issue of cosmic radiation, including its effects on airline crews, frequent flyer passengers, astronauts, and aircraft, satellites, and spacecraft equipment, is addressed in more than 30 pages, including a large number of tables and illustrative figures. The prehistory, history, and current aspects of metrology are presented in connection with their relation to NORM/TENORM/TENR. Environmental modeling regarding NORM/TENORM is briefly discussed, focusing on its objectives. We emphasize the need for sensitivity analysis of mathematical environmental models. Legal aspects of natural radiation are presented and discussed, including the current rather heterogeneous international approach. The need for harmonizing national and international regulatory frameworks to NORM/TENORM industries is pointed out, including guidance and practical implementation of existing concepts and principles. The potential of using natural radionuclides for making and using terrorist weapons is not ignored. Although for reasons of security, technical details are mostly absent, a discussion of this important current issue is presented, encompassing analytical aspects of the suitability of natural radionuclides already used for terror or homicide attempts. Illegal acquisition of natural radioactive materials, motivation, as well as modes that can be adopted for a terrorist attack with natural radionuclides, are discussed, as well as descriptions of what is usually called indistinctly *dirty bombs*. A risk assessment of radiological terrorism using natural radionuclides is presented as a guide to those involved directly or indirectly in impeding such terror attempts and attacks. A long list of references supports the statements, data, and overall information that appear in this book on TENR.

A book after being finished becomes a museum piece. However, people like Merril Eisenbud (1915–1997) and Konrad Bates Krauskopf (1910–2003) taught us that a book, like a museum, may be visited by those interested in learning from the past; teaching and undertaking research in the present; and developing new ideas, concepts, and research for the future. Thus, we feel a sense of achievement for making a little contribution to the vast book-museum that grows throughout the world every day.

We wish that this book will be helpful to students, scientists, and researchers in general, and to decision-making authorities an governmental and non-governmental organizations dealing with the NORM/TENORM/TENR issues. We hope also that officials in nuclear-, industrial-, aeronautical-, and space-agencies will find this book useful for making the critical assessment of the radiological impact of natural radiation vis-á-vis the radiological impact from man-made sources used in nuclear technology and nuclear medicine.

We owe gratitude to many colleagues in several countries who let us use their data, tables, and figures in this book. They are too many to be mentioned in this preface, but at least some of them are cited in the text. The TENR book could not have been written without their help.

In addition to the immense help of Murdoch Baxter, other members of the Elsevier staff helped us in different phases of the book project. We would like to mention at least those with whom we exchanged messages. They are the following: Linda Versteeg, Anita Koch, and Nicola Poser. More recently Mageswaran Babusivakumar made valuable suggestions and corrections to improve the quality of this book. All of the above-mentioned Elsevier staff members have helped at one time or another to overcome difficulties that looked at first glance unsurmountable. Our thanks are extensive to those Elsevier member staff who helped us *incognito* to put together this book.

Anselmo's wife, Alba, and his son, Claudio, provided permanent and necessary support to keep writing even when there were too many other commitments to attend to. We do have to confess that writing this book ends up being an enjoyable task.

Anselmo S. Paschoa
Saquarema, Rio de Janeiro, Brazil

Friedrich Steinhäusler
St. Gilgen, Salzburg, Austria
26 December 2009

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CHAPTER 1

INTRODUCTION

1.1. Is TENR a Universal Issue?

Natural radiation (NR) is a phenomenon that pervades the whole known Universe. The term radiation is usually applied only to electromagnetic radiation which includes radio waves, microwaves, infrared rays (thermal radiation), visible light, ultraviolet, X-rays, and gamma rays – covering 19 orders of magnitude in terms of wavelength or frequency. The spectrum of electromagnetic radiation, also known as the Maxwell spectrum, is schematically illustrated in Figure 1.1.

The expression technologically enhanced natural radiation (TENR) encompasses all kinds of NRs which are – or were at any time in the past – enhanced by technological activities. Thus, one can ask the following germane question – Is TENR a Universal issue?

The concept of Universe as mentioned here extends from a radius of 5.5 light hours (or less) for the solar system to about 15 billion light years for the observable Universe. The existence of the cosmic microwave background radiation (CMBR) was predicted some 60 years ago by George Gamow (Gamow, 1948a; Gamow, 1948b). Gamow suggested that the Universe was a sphere of radius *R* made of matter and radiation, with *R* expanding proportionally with time *t*. At about the same time, Ralph Alpher and Robert Herman estimated the temperature of the CMBR to be between 5 and 28 K (Alpher and Herman, 1948). Not until 1965 was the CMBR detected experimentally as electromagnetic radiation (Penzias and Wilson, 1965). The current estimate of the CMBR temperature is about 2.725 K (http://www.eso.org/public/outreach/press-rel/pr-2008/pr-13-08.html – accessed on July 23, 2008).

The three governing laws of radiation in the Universe are usually summarized today as the following:

1. Planck's law (black body) – which relates the radiant energy emitted by a given celestial body (object) to the inverse of the fifth power of the wavelength of the radiation and the temperature of the radiant object;
2. Stefan–Boltzmann law – which relates the total energy emitted by an object to the fourth power of temperature; and
3. Wien's law – which establishes that the peak radiance decreases linearly as the temperature increases.

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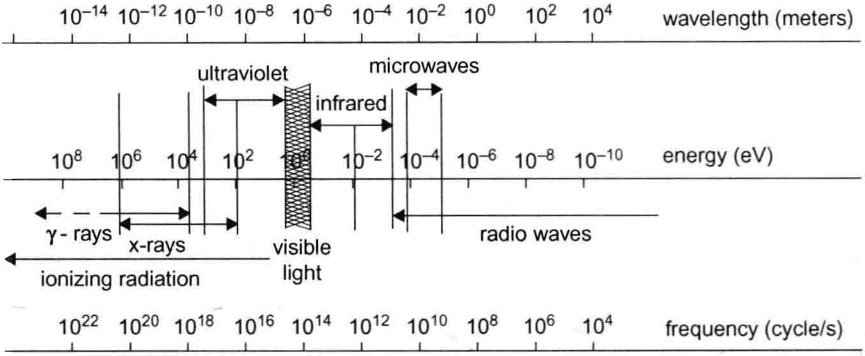

Figure 1.1 Electromagnetic spectrum or Maxwell spectrum.

When astrophysics was still considered a new scientific discipline, back in 1965, William A. Fowler was invited to give the fifth series of the Jayne Lectures at the American Philosophical Society in Philadelphia (Fowler, 1967). In his lectures, Fowler discussed the theoretical differences between the neutrinos and antineutrinos which appear in weak beta decay processes: for neutrinos, positron radioactivity, electron capture by positrons, and neutrino capture by neutrons; and for antineutrinos, electron radioactivity in the neutron decay process, positron capture by neutrons, and antineutrino capture by positrons.

The neutrino (and the antineutrino) had been postulated more than 30 years before Fowler's lecture in a letter written by Wolfgang Pauli to Lisa Meitner and Hans Geiger who were attending a meeting in Tübingen in December 1930 (Pauli, 1930). Pauli's letter preceded the discovery of the neutron by Chadwick by 2 years (Chadwick, 1932a, 1932b). The neutrino was finally detected by Frederick Reines and Clyde Cowan in 1956 (Reines and Cowan, 1956; Cowan et al., 1956).

The neutrino is a weak interacting particle with near-zero mass which travels with almost light speed. The very existence of the neutrino makes radiation a Universal issue.

Neutrinos are also produced on Earth either by natural radioactivity or in accelerators and nuclear reactors by the decays of natural radionuclides from the uranium isotope series (^{235}U and ^{238}U) and from the thorium (^{232}Th) series plus those from artificial radionuclides like plutonium and other transuranium isotopes. Neutrinos are also produced in the Sun and throughout the Universe in a myriad of nuclear reactions, including fusion.

There are three types of neutrinos ordered by increasing order of rest mass: electron-neutrino, muon-neutrino, and tau-neutrino. Some neutrinos detected on Earth are messengers from stars, including our Sun.

Neutrinos (i.e., geo-neutrinos) resulting from the decay chains of the ^{238}U, ^{235}U, and ^{232}Th plus the beta decay of ^{40}K are extant in the Earth's environment. The production of geo-neutrinos can be summarized by the series of simplified nuclear reactions as represented below:

$$^{238}U \rightarrow {}^{206}Pb + 8\,{}^{4}He + 6e^{-} + 6\bar{\nu}e$$
$$^{235}U \rightarrow {}^{207}Pb + 6\,{}^{4}He + 5e^{-} + 5\bar{\nu}e$$
$$^{232}Th \rightarrow {}^{208}Pb + 6\,{}^{4}He + 4e^{-} + 4\bar{\nu}e$$
$$^{40}K \rightarrow {}^{40}Ca + e^{-} + \bar{\nu}e$$

Several ongoing and future experiments are aimed at detecting geo-neutrinos (Bohem et al., 2001; SNO Collaboration, 2004; Fiorentini et al., 2005; de Meijer et al., 2006; Enamoto, 2006; Giammarchi, and Miramonti, 2006; Rubia, 2006; The Borexino Collaboration, 2006; Domogatsky et al., 2006; Learned, 2007; Raghavan, 2007; Neutrino Geoscience, 2008).

Neutrinos can also be used, however, in nuclear safeguards activities as a nonintrusive tool to check a reactor's activities (see, e.g., Bohem et al., 2001; Anjos et al., 2006; Bowden, 2008).

Figure 1.2 is an idealized graph of the rate of antineutrinos detected in (or thermal power of) a nuclear reactor as a function of time. The expected number of detected antineutrinos is of the order of a few hundred per day, depending on a number of parameters, which can be translated into the thermal power of the nuclear reactor.

Although one cannot say that humans can enhance the NR extant throughout the Universe to any significant degree, the Palo Verde, Double Chooz, and Angra dos Reis neutrino oscillations experiments show that nuclear reactors contribute a detectable number of electron–antineutrinos per fission of ^{235}U, ^{238}U, ^{239}Pu, and ^{241}Pu.

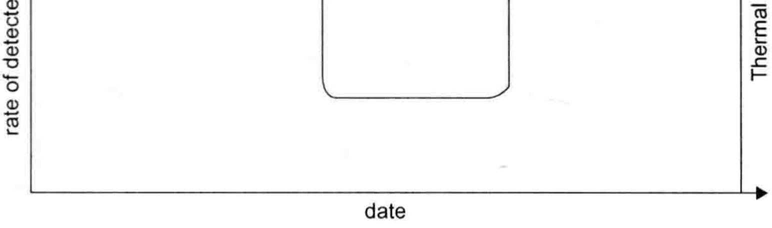

Figure 1.2 Idealized graph of the rate of antineutrinos detected in (or thermal power of) a nuclear reactor as a function of time.

Neutrinos are also produced artificially in nuclear detonations and particle accelerators (Zuber, 2004). Those neutrinos, like all other neutrino types mentioned above, are spread out all over but cannot be distinguished from those of natural origin.

Thus, one can say confidently that TENR is a Universal issue but, because the neutrinos interact very little with matter, the radiation doses from neutrinos do not need to be considered as far as radiation protection issues are concerned.

1.2. TENR – A GLOBAL ISSUE

The acronym TENR stands for technologically enhanced natural radiation, though the most commonly found definition in Internet dictionaries is "technologically enhanced natural radioactivity." The latter is a more restrictive definition derived from the wide use of the acronyms NORM (naturally occurring radioactive materials) and TENORM (technologically enhanced NORM).

As is well known, radioactive emissions encompass only the lower end of the Maxwell electromagnetic spectrum of radiation – as wavelength $10^{-15} > \lambda(\mathrm{m}) > 10^{-7}$ or as frequency $10^4 < \nu\ (\mathrm{Hz}) < 10^{22}$ – while radiation *per se* includes the whole Maxwell spectrum represented schematically in Figure 1.1. As one can see in Figure 1.1, the spectrum of visible light encompasses just a small portion of the Maxwell spectrum, corresponding to a narrow window from 400 to 700 nm in the middle of the wavelength spectrum, while gamma rays, for example, have wavelengths of 10^{-10} m. The Maxwell spectrum ranges from less than 10^{-15} m to more than 10^4 m. This book will address TENR as technologically enhanced natural radiation in a broader sense, but without emphasizing ultraviolet, visible, infrared, microwaves, and radio waves. In other words, this book will deal essentially with natural ionizing radiation as far as it is enhanced technologically.

One can confidently state that there are only two types of NR: cosmic radiation and terrestrial radiation. Some authors, however, do prefer to add internal sources of NR in the human body as a third type of radiation.

Here, it is worth mentioning that cosmic radiation (or cosmic rays) is defined in a pedestrian way as high-energy particles of extraterrestrial origin. A more classical definition states that cosmic rays are nuclei (or ionized atoms) which can be from a single proton up to an iron nucleus and beyond. Protons constitute 90% and alpha particles 9% of all cosmic rays, consequently *all other* components are less than 1%. *All other* means stable and quasi-stable particles like neutrons, antiprotons (and maybe anti-nuclei), hard gamma rays (i.e., $\lambda < 10^{-14}$ m), electrons, positrons, neutrinos, antineutrinos, esoteric particles like weakly interacting massive particles (WIMPS) – which are hypothetical particles assumed to be a possible solution for the dark matter problem (see, e.g., Feng and Kumar, 2008) – magnetic monopoles, mini black holes, etc. These are the primary cosmic rays (PCRs) and are produced in stellar flares (like solar flares), supernovae events, and in other extraterrestrial galactic and extragalactic energetic sources.

The primary cosmic ray spectra at 10.6 GeV per nucleon used in calculations are composed of protons in the hydrogen flux (the unbound or free protons) plus 11 groups of primary nuclei, as shown in Table 1.1.

The interactions between the PCRs and the atmospheric and terrestrial nuclei produce secondary cosmic rays (SCRs). The SCRs are essentially all elementary particles and stable and unstable nuclei from which the most important are the following: nucleons, nuclei and nuclides, hard gamma rays, mesons ($\pi^{\pm}$, π^{0}, $K^{\pm}$, …, $D^{\pm}$, …), charged leptons ($e^{\pm}$, $\mu^{\pm}$, $\tau^{\pm}$), and neutrinos and antineutrinos (ν_e, ν_μ, ν_τ).

Concerns for the potential radiation doses to crew and passengers of high-altitude supersonic commercial air transport due to cosmic rays from galactic and solar events started in the early 1960s (Foelsche, 1961; Foelsche and Graul, 1962). However, the interest in the radiation doses due to cosmic rays had begun in the 1950s (UNSCEAR, 1956 and 1958, WHO, 1959). There are not so many differences between the cosmic ray doses received by high-altitude populations and those received by crew and passengers of commercial airplanes flying at altitudes varying from 3 to 12 km. The latter data are now available through a large number of publications.

One of the first important actions of UNSCEAR was to carry out investigations on the biological effects of ionizing radiation (UNSCEAR, 1956, 1958). It was then accepted that the probability of mutation per *locus*

Table 1.1 Components of the primary cosmic ray spectrum at 10.6 GeV per nucleon (O'Brien, 2005).

Z	Element	Relative abundance	%
1	H	750	95.233
2	He	34	4.317
3–5	Li–Be	0.4	0.051
6–8	C–O	2.20	0.279
9–10	F–Ne	0.3	0.038
11–12	Na–Mg	0.22	0.028
13–14	Al–Si	0.19	0.024
15–16	P–S	0.03	0.004
17–18	Cl–Ar	0.01	0.001
19–20	K–Ca	0.02	0.003
21–25	Sc–Mn	0.05	0.006
26–28	Fe–Ni	0.12	0.015

per unit dose was of the order of 1×10^{-7}. As a consequence, it was also known that a rather large number of observations needed to be made to resolve the "*signal to noise ratio*" to obtain statistically significant results concerning the *dose response relationship*. Not long after the 1958 UNSCEAR report was published, a World Health Organization (WHO) Expert Committee also produced a report which expressed concern about the genetic effects which might be produced in humans due to the increasing use of ionizing radiation in medicine, science, and industry (WHO, 1959). It was then expected that the study of human populations exposed to relatively large amounts of background radiation (i.e., of the order of $1\,\mathrm{rem\,y^{-1}}$ – or $10\,\mathrm{mSv\,y^{-1}}$ in today's units) would bring untapped information on radiation-induced mutations and their fate.

As mentioned by Paschoa (2000a, 2000b), some of the obvious populations to be studied, in accordance with the WHO (1959) committee recommendations, were those living in high-altitude areas; for example, Cerro de Pasco, Peru, 4.3×10^3 m, latitude 10°S; the Himalayan area (Lhasa), 3.7×10^3 m, latitude 30°N; La Paz, Bolivia, 3.6×10^3 m, latitude 16°S; Quito, Ecuador, 2.9×10^3 m, latitude 0°; Bogota, Colombia, 2.6×10^3 m, latitude 4°N. However, areas with high natural radioactive background were also, in some cases, worth investigating. Among those, a WHO Committee mentioned the following (WHO, 1959): part of the Kerala State, in India, and adjoining area in Madras State; the monazite areas in the Brazilian States of Espírito Santo and Rio de Janeiro; the mineralized volcanic intrusives in the Brazilian States of Minas Gerais and Goiaz; the primitive granitic, schistous, and sandstone areas of France with slight elevation of NR; and some uninhabited areas of the Belgian Congo.

At that time, the doses from external and internal irradiation from natural sources under the usual conditions at sea level were believed to be as shown in Table 1.2 (WHO, 1959). The temporal evolution of concepts and the improvements of knowledge and measurement techniques resulted in new information on the doses from external and internal irradiation. The 1959 dose estimates did not have data on the inhalation exposure of either radon or thoron, though it had estimates for ^{14}C, because of the importance which was then given to the introduction of this radionuclide into the biosphere due to the nuclear weapon tests in the atmosphere (Suess, 1953; Revelle and Suess, 1957; Arnold and Anderson, 1957; Young et al., 1965). Ten years before the WHO Expert Committee met, ^{14}C had started being used as a reliable dating tool based on the ratio $^{14}\mathrm{C}/^{12}\mathrm{C} \approx 1.3 \times 10^{-12}$ found both in living organisms and in the atmosphere (Arnold and Libby, 1949; Libby, 1955). However, there was the suspicion after the atmospheric nuclear weapon tests that the carbon isotopic ratio would change to an extent that would affect the reliability of the method (see, e.g., Krane, 1987).

Table 1.2 Exposure rates from external and internal irradiation to gonads and other soft tissues, as known in 1959.

	Exposure rates	
	mrem y^{-1}	µSv y^{-1}
External irradiation		
Cosmic rays	28	280
Gamma rays (outdoors)	47	470
Internal irradiation		
^{40}K	19	190
^{14}C	1.6	16
^{226}Ra	?	?
Total (from all sources)	95	950

Source: Adapted from WHO (1959) – see also Paschoa (2000a, 2000b).

Table 1.3 compares as much as possible the dose estimates made by WHO (1959) with those made by UNSCEAR (2000). It is interesting to note, by observing the last column of Table 1.3, that though the dose estimates for cosmic radiation increased and ingestion exposure due to terrestrial radionuclides increased by only about 30% each, and the dose estimates from terrestrial external irradiation did not change significantly, the total dose estimates increased by about 150%. Radon (^{222}Rn) inhalation which was not accounted for in the WHO estimates in 1959 became by far the most important individual component of the dose estimates made by UNSCEAR in 2000.

The areas with high levels of natural radioactivity and/or high radon or thoron concentrations are likely to become TENR areas because of mineral exploitation and later industrial processing of NORM material, as has been pointed out in the Brazilian case (Paschoa, 2002). The WHO Committee (WHO, 1959) had suggested that populations exposed to annual doses of about 10 mSv (i.e., ≈4 times higher than the average dose to the world population), as per the UNSCEAR (2000) estimate, would bring untapped information on radiation-induced mutations and their effects. Much has been achieved as far as the dose assessment of elevated NR areas is concerned (see, e.g., Eisenbud, 1982; Sohabi and Esmaili, 2002). Occupational annual dose equivalents might have reached levels as high as 55 mSv from thoron inhalation prior to the improvement of industrial hygiene in a Brazilian monazite industry (Paschoa and Pohl-Rülling, 2005). Such high annual doses are more than 500 times the current estimate for the world average annual thoron inhalation dose, or approximately 20 times the world average dose from all sources.