DISSERTATIONES PHYSICAE UNIVERSITATIS TARTUENSIS
82

DISSERTATIONES PHYSICAE UNIVERSITATIS TARTUENSIS
82

MERLE LUST

Assessment of dose components to Estonian population

TARTU UNIVERSITY PRESS

The Dissertation was admitted on June 26, 2012 in partial fulfillment of the requirements for the degree of Doctor of Philosophy in physics and allowed for defense by the Scientific Council of the Institute of Physics, University of Tartu.

Supervisors: Dr. Enn Realo, Institute of Physics, University of Tartu

Dr. Rünno Lohmus, Institute of Physics, University of Tartu

Opponents: Dr. Raimo Mustonen, Radiation and Nuclear Safety Authority, Helsinki, Finland
Dr. Tõnu Viik. Tartu Observatory, Estonia

Defence: September 14, 2012 at University of Tartu, Tartu, Estonia

Publication of thesis was supported by: Graduate School on Functional Materials and Technologies (GSFMT), University of Tartu and Tallinn University of Technology, EU Social Funds project 1.2.0401.09-0079.

ISSN 1406–0647
ISBN 978–9949–32–082–0 (trükis)
ISBN 978–9949–32–083–7 (PDF)

Tartu Ülikooli Kirjastus
www.tyk.ee
Tellimus nr 370

CONTENTS

LIST OF PUBLICATIONS INCLUDED IN THE THESIS

I. Realo, K.; Isakar, K.; Lust, M.; Realo, E. (2007). Weekly variations of the ^{210}Pb air concentration in North Estonia. Boreal Environment Research, 12(1), 37–41.

II. Lust, M.; Realo, E. (2012). Assessment of natural radiation exposure from building materials in Estonia, Proceedings of the Estonian Academy of Sciences, 61, 2, 107–112

III. Lust, M.; Realo, E. (2012). Determination of Dose Rate from Chernobyl-derived Radiocaesium in Estonian soil. Journal of Environmental Radioactivity, in publishing.

LIST OF PAPERS NOT INCLUDED IN THE THESIS

IV. Lust, M.; Realo, K.; Realo, E. (2002). ^{137}Cs in soil profiles in NE Estonia. In: Proceedings of the 8th Nordic Seminar on Radioecology: The 8th Nordic Seminar on Radioecology, 25–28 Februar 2001, Rovaniemi, Finland. (Toim.) E. Ilus. Roskilde: Nordic nuclear safety research (NKS), 2002, (NKS; 70), 221–225.

V. Lust, M.; Realo, E.; Realo, K. (2003). Radiocaesium in NE Estonian soil. In: Radiation Protection in 2000's – Theory and Practice. Nordic Society for Radiation Protection. Proceedings of the XIII Ordinary Meeting: NSFS 13th Ordinary Meeting, Turku, 25–29 August 2002. (Toim.) W.Paile. Vantaa: Dark Oy, 2003, (STUK-A195; A195), 484–487.

VI. Realo, K.; Koch, R.; Lust, M.; Uljas, A.; Realo, E. (2004). Lead-210 in air and surface soil in NE Estonia. In: Full Papers of IRPA-11, 11th Intern. Congress of Intern. Radiation Protection Association: 11th International Congress of International Radiation Protection Association; Madrid, Spain; 23–28 May, 2004. Spain:, 2004, 1–8.

VII. Lust, Merle; Realo, Küllike; Realo, Enn (2006). Studies on Chernobyl radiocaesium in Estonia. Ympäristön radioaktiivisuus Suomessa – 20 vuotta Tshernobylista. Symposium Helsingissä 25.–26.4.2006, Helsinki. (Toim.) Ikäheimonen, T.K.. Helsinki: Dark Oy, 2006, (Säteilyturvakeskus; 217), 37–40.

VIII. Realo, Küllike; Isakar, Kadri; Lust, Merle; Realo, Enn (2006). Weekly variation of ^{210}Pb air concentration in North Estonia. In: STUK-A217: Ympäristön Radioaktiivisuus Suomessa – 20 Vuotta Tshernobylista; 25.–26.04.2006; Helsinki . Helsinki, Soome: Dark Oy, Vantaa, Soome, 2006, (STUK-A), 195–198.

IX. Realo, Küllike; Realo, Enn und Lust, Merle (1998). Eine Bewertung der Radiologishen Wirkung von Ölschifferflugasche in Estland. In Winter, M., Henrichs, K., Doerfel H. (Eds.). Radioaktivität in Mensch und Umwelt, Band II. IRPA/Fachverband für Strahlenschutz, Lindau, 867–872.

AUTHOR'S CONTRIBUTION

The papers that form the basis of this thesis are the result of collective work with important input and contribution from all of the co-authors with complementary expertise in different fields. The author's contribution to the papers can be described as follows:

- Paper I: The author was responsible for the collection of the air filter samples and participated in gamma-spectroscopy analysis of the samples. The author was responsible for writing the assessment part of the paper.
- Paper II: Coordination of sampling and experimental activities. The author carried out the majority of analysis of radionuclide concentrations in the building materials and prepared the computer code for calculating the doses to the public. Preparation of the overview of the topic based on the work with the scientific papers. The Author was responsible for the manuscript and figures.
- Paper III: Participation in the co-ordination of the sampling and taking part in majority of the samplings. The author determined radionuclide concentrations in the soil profiles and worked with the assessment tools. The Author was responsible for the manuscript.

ABBREVIATIONS AND SYMBOLS

BSS	–	Basic Safety Standards
Cm–V	–	Cambrian–Vendian aquifers
CTBTO	–	Comprehensive Nuclear-Test-Ban Treaty Organization
EC	–	European Comission
IAEA	–	International Atomic Energy Agency
ICRP	–	International Commission on Radiological Protection
UNSCEAR	–	United Nations Scientific Committee on the Effects of Atomic Radiation
WHO	–	World Health Organization
WSZ	–	Water Supply Zone

1. INTRODUCTION

The purpose of a radiation protection system is to protect people and the environment against the detrimental effects of radiation exposure without undue limitations on desirable human activities that may be associated with such exposure [1]. Three fundamental principles must be taken into account: justification, optimisation of protection and application of dose limits. Therefor dose values play an important role in a radiation protection system. However, in most cases doses to the public cannot be measured directly, or cannot be measured at all. This creates a need to use different methods to assess doses, based on model calculations or measurement data from environmental surveillance programs, or on their mix.

For an accurate assessment of the dose to a given population, several dose components have to be considered, for example – natural radiation, the use of radioactive material and medical exposure [2]. The dose due to ionizing radiation from natural sources constitutes the greatest contribution to the effective collective dose received on average by the world's population [3,4] and for decades, many countries have put a great deal of effort into quantifying irradiation dose rate levels [5,6].

In Estonia, there have been some studies performed in order to assess some of the dose to public components, like radon in Estonia [7]. However, in order to obtain an overall understanding, there is a need to assess all of the primary components. The general goal of this work is to estimate the dose components, which have not yet been assessed in Estonia. As a final result the overall estimation of the dose from environmental components will be presented.

This work includes both experimental and theoretical aspects. Gamma spectrometry was used in the estimation of the concentration of different radionuclides in a wide range of materials, such as soil samples, building materials and air filters. These measurement results were used in dose assessment. The work was performed over more than 10 years at the Institute of Physics, University of Tartu.

2. MOTIVATION AND AIMS OF THE RESEARCH

Several dose components for the Estonian population have previously been examined. However, for better estimation of the risks from ionizing radiation, it was important to attempt to estimate all the components of the dose. The aim of this study was to review different studies performed, and to establish the dose components that remained unestimated. During the study several components were estimated, so it was possible to estimate the dose to the Estonian population from the existing exposures.

More specifically the aims of the research were set out as follows:

1. Estimation of the seasonal variations and average values for ^{210}Pb concentration by using the filters from air sampling stations in Estonia.
2. Estimation of the dose component, which is caused by the ^{210}Pb concentrations in the air, taking account the local results of the average values and seasonal variations.
3. Analysis of the most common building materials with estimation of their activity concentrations, index *I* and *Ra (en)*.
4. Estimation of the doses due to the building materials based on the estimated radionuclide concentrations.
5. Estimation of local time-dependent migration and transfer rates of radiocaesium in soil using soil profiles, which were taken during the period 1991–2003.
6. Based on the estimated local depositions and the transfer rates, estimation of the 50 year effective doses caused by external gamma exposure from radiocaesium depositions in NE Estonia.
7. Estimation of the dose to the Estonian population from existing exposures.

3. BACKGROUND

The biological effects of radiation are caused mainly by their effect on living cells [8]. The body has repair mechanisms against damage induced by radiation as well as by chemical carcinogens; therefore for low levels of radiation exposure, the biological effects are so small they may not be detected.. The effects of low doses of radiation, if any, would occur at the cellular level, and thus changes may not be observed for many years (usually 5–20 years) after exposure [8].

In addition to the radiation doses caused by natural sources, members of the public can be exposed to radiation through effluent discharges with some levels of radioactivity, from materials released from nuclear facilities, from the transport of radioactive material to and from facilities, and from the management of radioactive waste [1, 9]. Doses to the public can be a result of the direct external exposure to radiation associated with radioactive materials, or from internal exposure due to intake of radionuclides in air, foodstuffs and water. The population can also be exposed to radiation through accidents in which control is lost over a radiation source within a facility or in which radioactive material is released into the environment. Such releases can provide both air and water borne exposure. Accidental releases generally take place over short time periods, which can last from a few hours to days.

In all situations involving exposure to radiation, an appropriate level of radiation protection needs to be applied using a graded approach, i.e. the level of control applied must be commensurate with the associated risk or risks. Exposure to radiation is defined in terms of: planned exposure situations, emergency exposure situations and existing exposure situations [10]. A planned exposure situation arises from the planned operation of a source or from a planned activity that results in an exposure from a source. An emergency exposure situation arises as a result of an accident, a malicious act, or any other unexpected event, and requires prompt action in order to avoid or reduce adverse consequences. An existing exposure situation is a one of exposure which already exists for which a decision regarding control needs must be taken [1]. Examples of an existing exposure situation are exposure to natural background radiation and exposure to residual radioactive material from a nuclear or radiological emergency after the emergency exposure situation has been declared ended. Several studies have proved that the component of exposure due to natural background sources is largest [3, 4, 5, 6]. There are many types of existing exposure situations that may cause exposures high enough to initiate radiological protective actions, or at least their consideration. Lately there has been a great deal of discussion about regulating radon in dwellings in European Union and also Estonia, mostly due to the exposure concerns.

In assessment of doses the absorbed dose is used as the fundamental physical quantity [1] given by

$$D = \frac{d\varepsilon}{dm},$$

where $d\varepsilon$ is the mean energy imparted to matter of mass dm by ionising radiation. The SI unit for absorbed dose is joule per kilogram (J kg^{-1}) and its special name is gray (Gy).

Equivalent dose is the dose in a tissue or organ T given by:

$$H_T = \sum_R w_R D_{T,R},$$

where $D_{T,R}$ is the mean absorbed dose from radiation R in a tissue or organ T, and w_R (Table 1.) is the radiation weighting factor. Since w_R is dimensionless, the unit for the equivalent dose is the same as for absorbed dose, J kg^{-1}, and its special name is sievert (Sv).

Table 1. Radiation weighting factors recommended by ICRP [1]

Radiation type	**Radiation weighting factor w_R**
Photons	1
Electrons and muons	1
Protons and charged pions	2
Alpha particles, fission fragments, heavy ions	30
Neutrons	A continuous function of neutron energy

The effective dose is the tissue-weighted sum of the equivalent doses in all specified tissues and organs of the body, given by the expression:

$$E = \sum_T w_T \sum_T w_R D_{T,R},$$

where w_R $D_{T,R}$ is the equivalent dose in a tissue or organ, T, and w_T (Table 2.) is the tissue weighting factor. The unit for the effective dose is the same as for absorbed dose, J kg^{-1}, and its special name is sievert (Sv).

Table 2. Tissue weighting factors recommended by ICRP [1]

Tissue	**Tissue weighting factor w_T**
Bone-marrow (red), Colon, Lung, Stomach, Breast, Remainder tissues	0.12
Gonads	0.08
Bladder, Oesophagus, Liver, Thryroid	0.04
Bone surface, Brain, Salivar glands, Skin	0.01

The procedure for the assessment of the effective dose adopted by ICRP is to use the absorbed dose as the fundamental physical quantity, average it over specified organs and tissues, then to apply suitably chosen weighting factors to take into account differences in the biological effectiveness of different radiations in order to give the quantity equivalent dose. To consider differences in sensitivities of organs and tissues weighted for the radiosensitivity of these organs and tissues. The effective dose is based on the exposure to radiation from external and internal exposures, as well as on the primary physical interactions in human tissues and judgements regarding biological reactions resulting in stochastic health effects [1]. The effective dose is used in radiation protection for planning purposes and the optimisation of protections for occupational workers and the general public. It is also utilised for demonstrating compliance with dose limits and in this sense, effective dose calculations are used for regulatory purposes worldwide.

An assessment can be prospective or retrospective. Prospective doses are estimated for individuals whose exposure has not yet occurred, while retrospective doses are generally estimated for groups of individuals that are known to have received exposures [9]. The dose assessment process is described in Fig 1.

Dose limits apply only in planned exposure situations. For occupational exposure in planned exposure situations, the International Commission on Radiological Protection recommends 20 mSv per year, averaged over defined 5 year periods (100 mSv in 5 years), with the further provision that the effective dose should not exceed 50 mSv in any single year [1]. For public exposure in planned exposure situations it is recommended that the limit should be expressed as an effective dose limit of 1 mSv in a year. These limits are also enforced by Estonian legislation [11]. The limits on effective dose apply to the sum of doses due to external exposures and committed doses from internal exposures due to intakes of radionuclides. There are no legislative limits set for existing exposure situations.

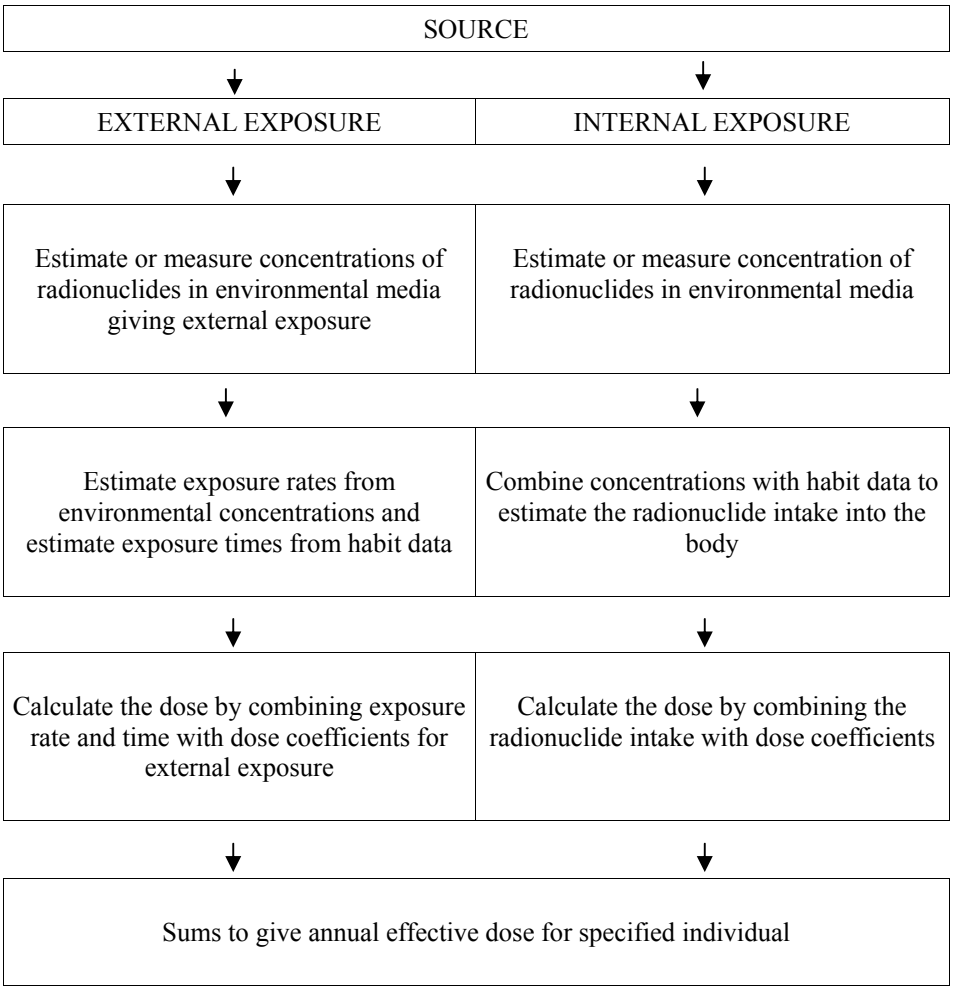

Figure. 1. Dose-assessment process

4. DOSE AND ITS COMPONENTS

The annual dose in millisieverts (mSv) due to natural sources of radiation, as estimated by UNSCEAR [4], are summarized in Table 3.

Table 3. Annual doses due to natural sources [4].

Source or mode	Annual average dose (mSv)	Typical range of annual dose (mSv)	Comments
Inhalation (radon gas)	1.26	0.2–10	Dose is much higher in some dwellings.
External terrestrial	0.48	0.3–1	Dose is higher in some places.
Ingestion	0.29	0.2–1	
Cosmic radiation	0.39	0.3–1	Dose increases with altitude.
Total natural	2.4	1–13	Sizeable population groups receive 10–20 mSv annually.

There is a high rate of variance in annual exposures. A study done in the UK [5] came to the conclusion that the average annual dose from natural radiation was 2.23 mSv, and about half of this resulted from indoor radon exposure. Artificial sources of radiation are subject to variations and trends that reflect current technology and radiological protection practices. The average annual dose from artificial radiation was found to be a little over 0.42 mSv, and mainly derived from the use of X-rays in medical procedures. The overall average annual dose is therefore almost 2.7 mSv. In the USA, exposure from natural and artificial sources is almost the same, 1.55 mSv per year, which means that the annual dose is estimated to be 3.1 mSv [6]. In order to have some understanding regarding the dose range, Table 4 below presents a selection of average doses received from artificial sources of radiation:

Table 4. Examples of the doses from artificial sources in comparison with those from natural sources [4].

Source or mode	Typical dose (mSv)
10 hour aeroplane flight	0.03
Annual dose from natural background	2.4
Annual dose to nuclear worker	1
Chernobyl recovery workers in 1986	150

Below, the following components of the dose and their estimated contibution to the annual exposure of the Estonian population are described:

- Radon
- Terrestrial radionuclides and cosmic radiation
- Radionuclides in foodstuff and drinking water
- Electricity production from oil-shale
- Building materials
- Atmospheric nuclear testing and Chernobyl accident fallout
- Medical exposure

Additionally to these doses, there are also workers who are involved in radiation practices and will recieve additional doses. According to the data on the doses of 1272 radiation workers in Estonia, the average occupational dose was 1.20 mSv in 2007 [12]. Based on the field of activity, the average annual doses are the following:

- Medicine: 1.36 mSv, number of radiation workers 1049, number of organizations 68;
- Industry: 1.03 mSv, number of radiation workers 223, number of organizations 22.

4.1. Radon

The inhalation component of an annual dose exposure is caused mainly by radon and its progeny. Radon-222 (^{222}Rn) is a radioactive decay product of uranium (^{238}U) which is present in the earth's crust in varying concentrations [7]. Gaseous radon is capable of moving from the soil to indoor environments via air currents. Penetration is dependent on the type of building and/or location. Radon isotope ^{220}Rn is a radioactive decay product of thorium (^{232}Th) that is also present in the earth's crust. Both ^{222}Rn and ^{220}Rn may also be emitted from some building materials. The concentration of radon in buildings may vary in the range of several orders of magnitude.

As radon is inert, nearly all of the gas inhaled is subsequently exhaled. However, when inhaled, the short-lived radon progeny can be deposited within the respiratory tract. Depending on the diffusion properties of the particles, the decay products present in the air can deposit in the nasal cavities, on the walls of the bronchial tubes and in the deep lung. Two of these short-lived progeny, polonium isotopes ^{218}Po and ^{214}Po, emit alpha particles and the energy deposited by these high-energy alpha particles may lead to health effects, principally lung cancer. Epidemiological evidence indicates that indoor radon is responsible for a substantial number of lung cancers in the general population[13]. Radon gas is by far the most important source of ionizing radiation among those that are of natural origin.

Surveys have been carried out to determine the distribution of residential radon concentrations in most European countries [14]. The worldwide average indoor radon concentration has been estimated at 39 Bq/m^3 [3, 15,16, 17]. The proportion of all lung cancers linked to radon is estimated to lie between 3% and 14%, depending on the average radon concentration in the country and on the method of calculation [13]. A recent study [18] has shown that radon in homes causes about 20,000 lung cancer deaths in the European Union (EU) each year. This is about 9% of the total lung cancer deaths in the EU and about 2% of cancer deaths overall.

The systematic mapping of radon levels in Estonia began in 1994. Since then, several additional projects have been carried out to measure radon levels in soil, air and in dwellings. In Estonia, the main source is indoor radon: radon emitted from the soil, its progeny attached to airborne particles and transported indoors via air movement. Building materials with enhanced radium concentrations are not known in Estonia [7]. Usually, the highest indoor radon concentrations have been found in the northern part of Estonia where uranium rich Dictyonema shale and uranium containing phosphorous Glauconite sandstone layers exist both in the bedrock and as fragments in the surface soils. Radon concentrations higher than 400 Bq m^{-3} have also been measured in buildings situated in areas with karst formations. Areas with Dictyonema shale, Glauconite sandstone and karst are areas with a special risk for radon.

Based on the measurements performed in 1994–2001, the map of the national average radon levels was prepared and published (Fig. 2) [19]. The map of radon concentrations indoors was prepared by the Estonian Radiation Protection Centre (currently known as The Environmental Board) in co-operation with Swedish colleagues [7, 20]. A radon risk map of Estonia is also available, prepared by the Estonian Geological Survey in co-operation with Swedish colleagues [21].

Taking into account the average radon concentrations in dwellings and the diversity of different building types, the dose component from radon was assessed. It was taken into account that the majority of the Estonian population lives either in a detached house or an apartment building. Based on these results it was estimated that indoor radon provides in Estonia an average annual dose of 1 mSv [7].

New epidemiological data is available about health effects caused by radon – on the assumption of an equilibrium factor for ^{222}Rn of 0.4 and an annual occupancy rate of 2000 hours, the value of activity concentration due to ^{222}Rn of 1000 Bq m^{-3} corresponds to an annual effective dose of the order of 10 mSv. Based on this ICRP recommends [1] using higher value of the dose conversion factor of 12.5 nSv/(h Bq m^{-3}) in the assessment of doses caused by radon and this was the reason for the reassessment of the above doses. Using the new factor the annual average dose to Estonian populations is estimated to be 2.1 mSv.

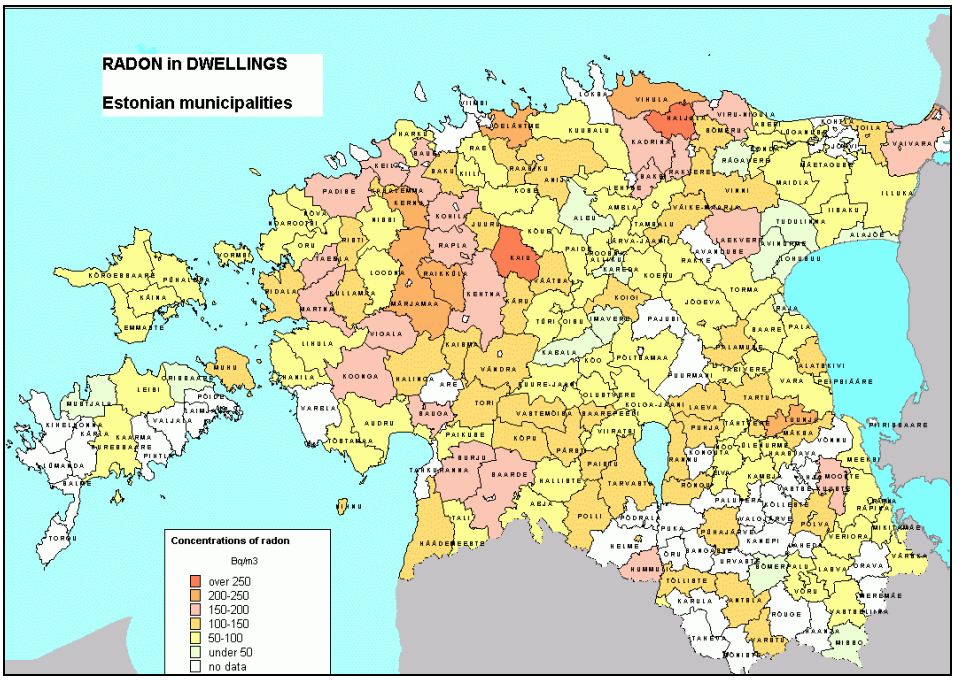

Figure 2. Estonian indoor radon map [19].

4.2. Terrestrial radionuclides and cosmic radiation

4.2.1. External

Most radionuclides in the uranium and thorium series, and ^{40}K, emit gamma radiation. A world-wide review [3] of the concentrations of ^{40}K, ^{238}U and ^{232}Th in soil gave median values of 400, 35 and 30 Bq kg^{-1}, respectively. The radionuclides, which are present in soils and other natural minerals, give rise to exposure from gamma radiation outdoors.

Primary cosmic radiation consists of very energetic charged particles moving through space. They originate mostly from events beyond our solar system, with a lower energy component originating from the Sun. The intensity of cosmic radiation is affected by the Earth's magnetic field, and there is also some variation of the dose rate with latitude. UNSCEAR [3] has produced an estimate of the cosmic radiation dose rate.

Based on the correlations of naturally occurring radioactive materials (NORM), activity concentrations confirmed the existence of at least two general soil types in Estonia, characterized by two different concentration ratios, C(U)/C(Th). While in the majority of the territory the concentration of U is approximately equal to that of Th, in the coastal region of N Estonia and especially of NE Estonia, significantly above-average uranium concentrations up to

350 Bq kg^{-1} are accompanied by the elevated C(U)/C(Th) ratios in the range of 1.5 … 5.

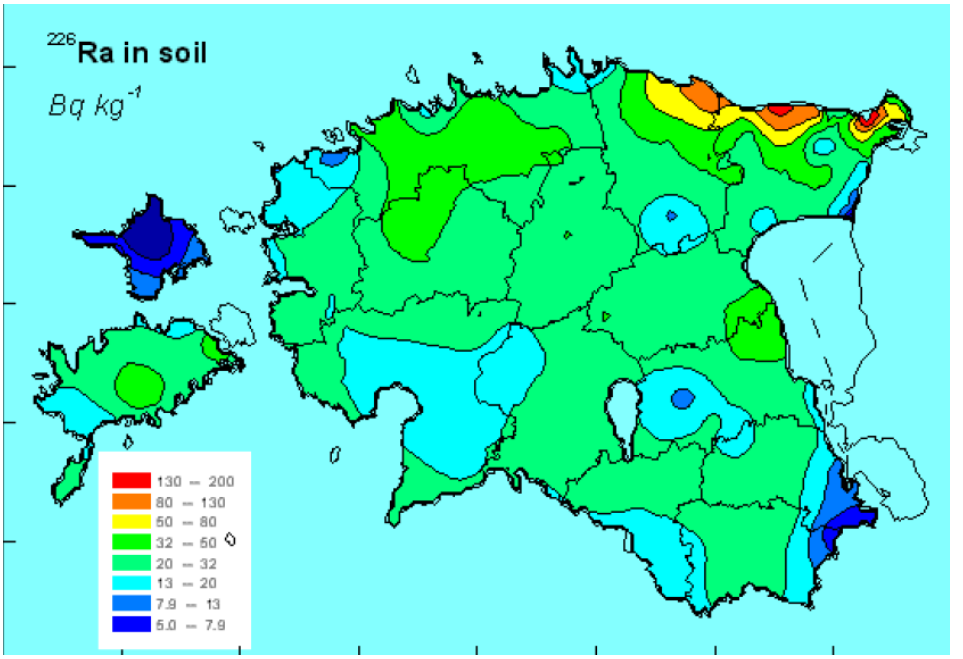

Figure 3. Estimated ^{226}Ra activity concentrations in Estonia [22].

Based on the study of activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in Estonian soils [22], the dose rates from terrestrial radiation were estimated. The methodology and dose coefficients used in the assessment were based on previous studies [3, 23]. The average annual effective dose of 0.80 mSv (with the range from 0.43 to 2.38 mSv), assuming 100% outdoor exposure time, from terrestrial radiation was estimated. The results take into account also the cosmic radiation component of 35 nSv/h (0.31 mSv/a).

Based on this data it is possible to estimate the annual dose to the members of the Estonian population from terrestrial radionuclides and cosmic radiation. The amount of time people spend inside buildings must also be taken into account. According to the UNSCEAR estimation [3] the indoor occupancy fraction is 0.8, which means people spending time outdoors about 20% of the time. Using these assumptions, the annual outdoor dose component caused by terrestrial radionuclides is in the range of 0.09 to 0.48 mSv, with the average of 0.16 mSv per year, in Estonia [4].

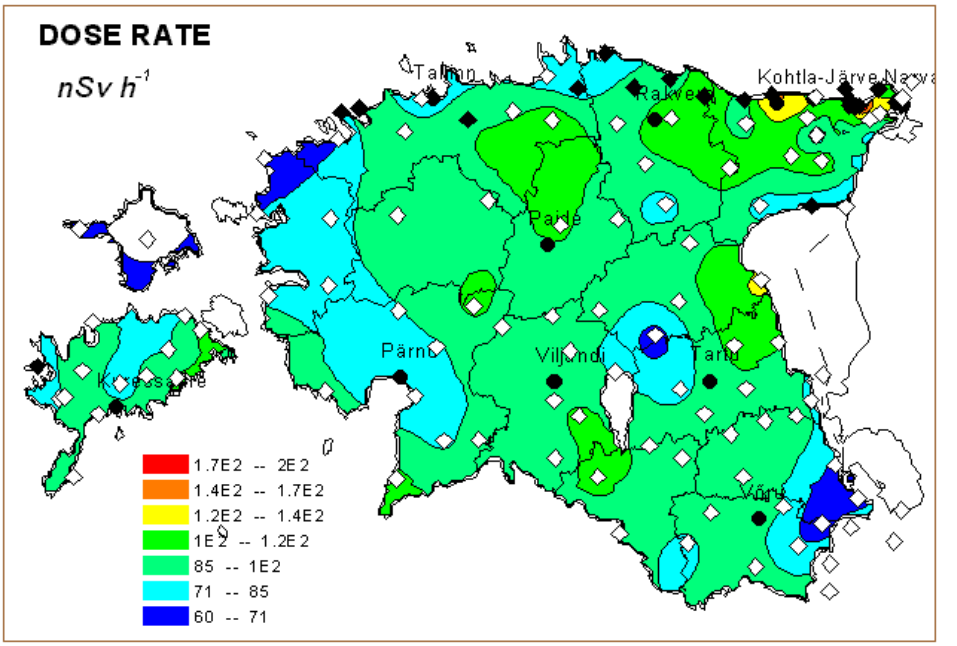

Figure 4. Estimated external dose rates in Estonia (terrestrial radionuclides + cosmic radiation) [22].

4.2.2. Internal

The subgroup ^{210}Pb–^{210}Bi–^{210}Po is responsible for an important contribution to human internal exposure. ^{210}Pb is a product of the ^{238}U decay series. It is derived from the decay of gaseous ^{222}Rn, the daughter of ^{226}Ra [24]. ^{226}Ra occurs naturally in soils and rocks and diffusion of a small proportion of the ^{222}Rn from the soil introduces ^{210}Pb into the atmosphere. Long-lived ^{210}Pb as a favourable tracer is widely used in environmental research, e.g., in the dating of lake sediments and ice cores; for the determination of sediment accumulation rates; in atmospheric aerosol and pollutant research; and in the modelling of atmospheric transport and removal processes. As the continental and marine radon source rates differ by about two orders of magnitude, the global as well as regional and local meteorological conditions strongly influence the ^{210}Pb surface air concentration. In the marine-to-continental, polar-to-temperate and temperate-to-equatorial interface regions these influences are especially significant [25].

Lead-210 is characterized by a complex behaviour in the environment. Radioactive ^{210}Pb with a relatively long lifetime ($t_{1/2}$ = 22.3 a) forms from its parent ^{222}Rn ($t_{1/2}$ = 3.82 d), which is a daughter product from the α-decay of ^{226}Ra. A minor radium impurity occurs in almost all soils, minerals and in water. In soil, α-recoil of radium releases some noble gas radon atoms from mineral grains into the pore space of soil, while the others remain fixed in soil particles.

The released radon atoms are transported by advection and diffusion through the pores and a fraction of them is released into the atmosphere. The worldwide average flux of ^{222}Rn to the atmosphere is evaluated 15–22 mBq·m^{-2}·s^{-1} from continental soils and 0.2 mBq·m^{-2}·s^{-1} from ocean waters [26]. Both released and fixed radon nuclei undergo a number of decays to produce ^{210}Pb. Air concentrations of ^{210}Pb determined by different authors vary in a wide range from 100 to 2500 μBq·m^{-3} with the worldwide average of 500 μBq·m^{-3} [3]. Its residence time in the atmosphere is considered to be relatively short, from one to two weeks [26, 27] and significant seasonal, temporal and geographical variations of air concentration are observed. Precipitation, predominantly in the form of rain and snow, scavenges ^{210}Pb from the atmosphere and governs a reasonably constant annual depositional flux to the soil. The long-term accumulation compensates losses caused by radioactive decay and migration to deeper soil layers. As a result, higher concentration of ^{210}Pb in comparison with ^{226}Ra is usually observed in the surface layers of soil. In these soil stratums, the total concentration of ^{210}Pb consists of two components: the atmospherically derived unsupported fraction and the *in-situ* produced supported fraction. Inventories of the unsupported fraction in soil can be related to the average annual atmospheric ^{210}Pb deposition fluxes. An estimation of the Estonian dose component from ^{210}Pb is given in chapter 6.1.

4.3. Foodstuffs and drinking water

Natural radionuclides in the environment may be taken up by plants and hence enter into the human food chain. Average annual doses are calculated using the average foodstuff intake and the reported radionuclide concentrations in those foodstuffs. In Estonia, monitoring of radionuclides in foodstuffs is carried out as part of the radiation monitoring program [28]. It includes examining food samples in two hospitals twice annually. In the analysis the following radionuclide concentrations are estimated: ^{137}Cs, ^{90}Sr and ^{40}K. Based on the data of the 2010 measurements, the assessment done by the Environmental Board concluded that the dose component from foodstuffs is about 0.2 mSv per year. This dose is primarily attributable to ^{40}K, the dose component caused from artificial radionuclides ^{137}Cs and ^{90}Sr was estimated to be less that 0.0006 mSv [28].

The presence of small amounts of radionuclides in drinking water is a common natural phenomenon. Since radium isotopes are more radiotoxic than uranium isotopes, significant exposure may occur when their concentration is relatively low.

In Estonia there are four superposed groundwater tables called the Quaternary, Silurian, Ordovician and Cambrian–Vendian (Cm–V) aquifers, according to their geological origin. Several studies have been carried out in order to estimate the dose caused by the drinking water in Estonia. A great deal

of analytical data on waterworks, especially in Northern Estonia, were available, albeit rather inhomogeneous. Upon first examination, they showed that 92% of the Water Supply Zones (WSZs) served by the Cm-V aquifer – vs. 21% of WSZs served by other aquifers – exceeded the 0.1 mSv/year value. The WSZs fed by the Cm-V aquifer are 140 of 912 in the country, serving roughly 250,000 people, or 22% of Estonian population. Specifically, 97% of available data regarding Cm-V supplied water resulted in doses higher than 1 mSv/year in, ranging as high as 12 mSv/year. The average dose component for the Estonian population from Cm-V aquifer supplied drinking water was estimated at 0.3 mSv [29], with the range of 0.02 – 0.95 mSv [30, 31].

4.4. Electricity production

Electricity production in Estonia is based mainly on the use of oil-shale in two large power plants in the North East. There is currently an increased interest in measuring naturally occurring radioactive materials (NORM) in power generation sources like coal, oil-shale and fly ash considering the health hazards caused by naturally occurring radionuclides. Oil-shale, like most fossil fuels, contains ^{238}U, the most important parent element of the natural uranium decay series. Large quantities of fly ash are produced by thermal plants and they may contain enhanced levels of radionuclides, along with other toxic elements [32]. The activity concentrations of the radionuclides in oil-shale ash were estimated [33].

Table 5. Activity concentrations in oil-shale ash [33].

Radionuclide	**Activity concentration (Bq kg^{-1})**
^{210}Pb	102±4
^{226}Ra	61.1±0.4
^{238}U	64±3
^{235}U	3.0±0.9
^{232}Th	29.5±0.6
^{40}K	1103±20

In the study the modelled ^{226}Ra deposition results were used for a comparison with the measured ones [34]. The depth-dependent activity concentrations of natural radionuclides in soil samples, taken from the vicinity of oil-shale power plants in NE Estonia, were determined by gamma spectrometry. As a result of the long-term operation of these power plants and the deposition of fly-ash in the vicinity, an increased concentration of natural radionuclides were found to be present in the surrounding air and surface soil.

In the previous study a MATLAB-based software package was designed and used for modelling of the long-term atmospheric dispersion and the geographical distribution of the deposited oil-shale fly-ash radionuclides [35]. Annual individual doses via external exposure, inhalation and ingestion pathways from oil-shale-based energy production have been calculated as well as the deposition amounts of fly-ash radionuclides in the surroundings of power plants. Total annual effective doses reach values up to 0.02 mSv.

4.5. Building materials

Naturally occurring radionuclides in the building materials may be a significant exposure component of the annual exposure dose. The worldwide average indoor effective dose due to gamma rays from the building materials is estimated about 0.4 mSv per year [36, 37]. Elevated indoor external dose rates may arise from high radionuclide content in building materials [38, 39, 40, 41, 42]. Generally, natural building materials reflect the geology of their site of origin. The average activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in the Earth's crust are 35 Bq kg^{-1}, 30 Bq kg^{-1} and 400 Bq kg^{-1}, respectively. However, elevated levels of natural radionuclides causing annual doses of several mSv were identified in some regions around the world, e.g. in Brazil, France, India, Nigeria, Iran [3, 36, 37].

Table 6. Annual effective dose (mSv) for natural radionuclides sources in concrete calculated from mean activity concentrations for selected countries [3].

Country	**Total annual effective dose (mSv)**
Greece	0.39
Egypt	0.41
Algeria	0.44
Swizerland	0.49
Russian Federation	0.49
United States	0.53
India	0.69
Sweden	0.78
Norway	0.86

The ALARA principle for building materials (the dose exposure indoors should be as low as reasonably achievable) is followed by using the index, I_{ext}. In a general case of a number x of different building materials in a room, the index is calculated as [43, 44]:

$$I_{ext} = \sum_{x} \frac{\sum_{i=1}^{n} w_{mi} C_{x_i}}{A_x}$$

where x represents the nuclide of interest, n is the number of different kinds of buildings materials used in a room, C_{xi} (Bq kg^{-1}) is the measured activity of each nuclide in the building material, w_{mi} is the weight fractional usage of the building material i and A_x (Bq kg^{-1}) is the parameter value representing the activity concentration of each nuclide of interest, which promoted an effective dose of 1 mSv per year.

For a specific building material the parameters, A_x, have the following values: 300 Bq kg^{-1} for ^{226}Ra; 200 Bq kg^{-1} for ^{232}Th and 3000 Bq kg^{-1} for ^{40}K. Accordingly, this index is defined by international as well as by Estonian legislation [11, 44] as the following sum:

$$I = \frac{C_{Th}}{200 Bq/kg} + \frac{C_{Ra}}{300 Bq/kg} + \frac{C_K}{3000 Bq/kg}.$$

where C_{Th}, C_{Ra} and C_K represent mean activity concentrations (Bq kg^{-1}) of the following radionuclides: ^{232}Th (in equilibrium with its daughter nuclides), ^{226}Ra (in equilibrium with its daughters) and ^{40}K.

Activity indexes are used to assess whether the safety requirements are being fulfilled. For construction materials the activity indexes are calculated on the basis of the measured activity concentrations of radium (^{226}Ra), thorium (^{232}Th) and potassium (^{40}K). In special cases other nuclides are also considered, e.g. caesium (^{137}Cs) in fuel ash of peat or wood. If the value of the activity index is 1 or less, the corresponding material can be used without restrictions as regards radioactivity. If the value exceeds 1, the responsible party (producer or dealer) is required to assess the radiation exposure caused by the material and show specifically that safety requirements are fulfilled.

The guidance for construction materials does not usually include possible radon releases to the indoor air from building materials. The values of the activity concentration index depend on the dose criterion and the mode and the quantity of material used in a building. According to the recommendations by the European Commission [44] for materials used in bulk amounts, the activity index should be less than 0.5. However, for superficial and other materials with restricted use the corresponding activity index should be between 2 and 6.

The radium equivalent concept allows a single index or number to describe the gamma output from different mixtures of uranium (i.e., radium), thorium, and ^{40}K in a material. The radium equivalent Ra(eq) in Bq kg^{-1}can be calculated as follows [43, 45]:

$$Ra\ (eq) = A\ (Ra) + 1.43*A\ (Th) + 0.077*A\ (K)$$

where *A (Ra)* is the activity of ^{226}Ra (which is usually the same as that of ^{238}U) in Bq kg^{-1}, *A (Th)* is the activity of ^{232}Th in Bq kg^{-1} and *A (K)* is the activity of ^{40}K in Bq kg^{-1}

Large-scale surveys of the concentration of radioisotopes in construction materials were summarized by the United Nations Scientific Committee on the Effects of Atomic Radiation, UNSCEAR. The European Union has established a database of activity concentration measurements of natural radionuclides (^{226}Ra, ^{232}Th and ^{40}K) in building materials. It contains about 10 000 samples of both bulk material (bricks, concrete, cement, natural- and phosphogypsum, sedimentary and igneous bulk stones) and superficial material (igneous and metamorphic stones) used in the construction industry in most of the European Union Member States [46].

Table 7. ^{226}Ra, ^{232}Th and ^{40}K activity concentrations in European brick, concrete, cement, natural gypsum: comparison with the European soil. Average values and ranges [46].

Building material	**Number of samples**	**Mean (and range) activity concentration (Bq kg^{-1})**		
		^{226}Ra	**^{232}Th**	**^{40}K**
Brick	1676	47 (2–148)	48 (2–164)	598 (12–1169)
Concrete	2727	60 (1–1300)	35 (1–152)	392 (7–1450)
Cement	2013	45 (4–422)	31 (3–266)	216 (4–846)
Natural gypsum	502	15 (1–70)	9 (1–100)	91 (5–279)
Soil	Number of countries			
European member state average	23	36 (0–1000)	34 (1–258)	483 (0–3200)

Taking into account the wide range of the radionuclide concentrations in the building materials, it is easy to understand that the index I do vary in wide range. An example of the range for the index I in bricks (1593 samples from EU countries) is given in Figure 5 [47].

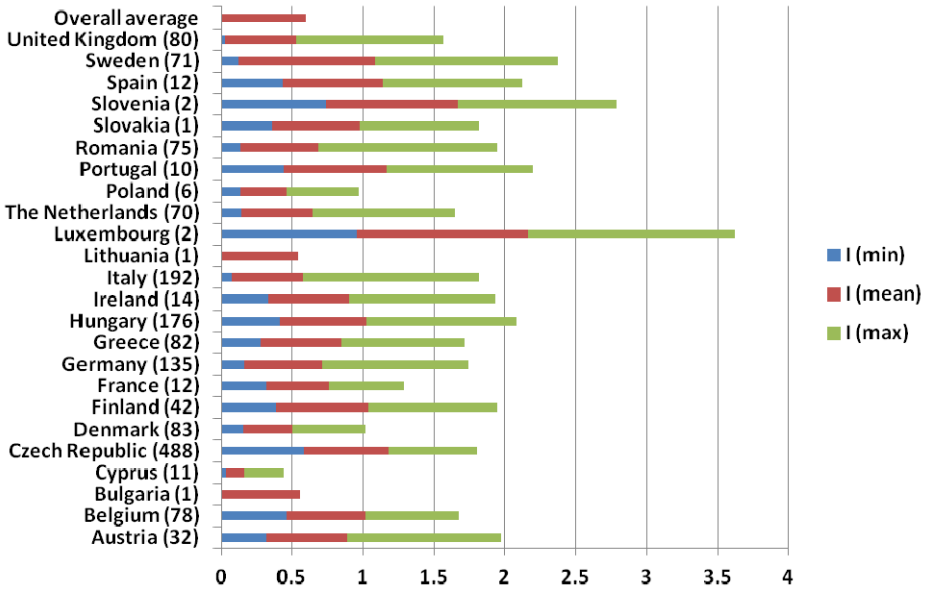

Figure 5. Mean and range of Index I in bricks [47].

The estimated annual doses due to building materials in Estonia are given in Chapter 6.2.

4.6. Radiocaesium fallout

Radiocaesium is a man-made radionuclide associated with nuclear fission facilities and the testing of nuclear weapons, and its deposition from the atmosphere has exhibited significant temporal variation. Radiocaesium contamination in the soil was produced by two major fallouts: a global fallout caused by nuclear weapons testing (NWT) in the atmosphere (1950s to 1970s) and a regional one – during and after the Chernobyl accident in 1986.

It was realised at an early stage that testing of nuclear weapons in the atmosphere would lead to some dispersion of radioactive particles [48, 49]. There were a fractional number of particles that penetrated the stratosphere and deposited worldwide over a period of many months. The explosive yield of bombs can be used as an indicator of the atmospheric behaviour of the debris. Radionuclides from bombs of less that 100 kilotons tend to remain in the troposphere, whereas stratosphere injection is almost complete for detontations larger than 500 kt [50].

The first test explosion of a nuclear weapon which left atmospheric traces was in a New Mexico desert in the U.S. in July 1945. More than 500 such tests were conducted until a ban become mostly effective in 1962 [3, 50]. The total number of detonations (including Hiroshima and Nagasaki) is 543 [51].

In the Northern hemisphere, global fallout commenced in the early 1950s, with a maximum deposition rate reached in 1963, the year of the most intensive nuclear weapons testing [52]. Since the late 1970s, rates of ^{137}Cs deposition have been very low, although there was an additional short-term input of radiocaesium in 1986, caused by the Chernobyl reactor accident.

The areal distribution of the ^{137}Cs fallout after the Chernobyl accident was extremely uneven in Estonia (Fig. 6). Maximum deposition occurred in NE Estonia, where in a few locations soil concentrations over 40 kBq m^{-2} were recorded, while the country-wide mean was 2.0 kBq m^{-2} [53].

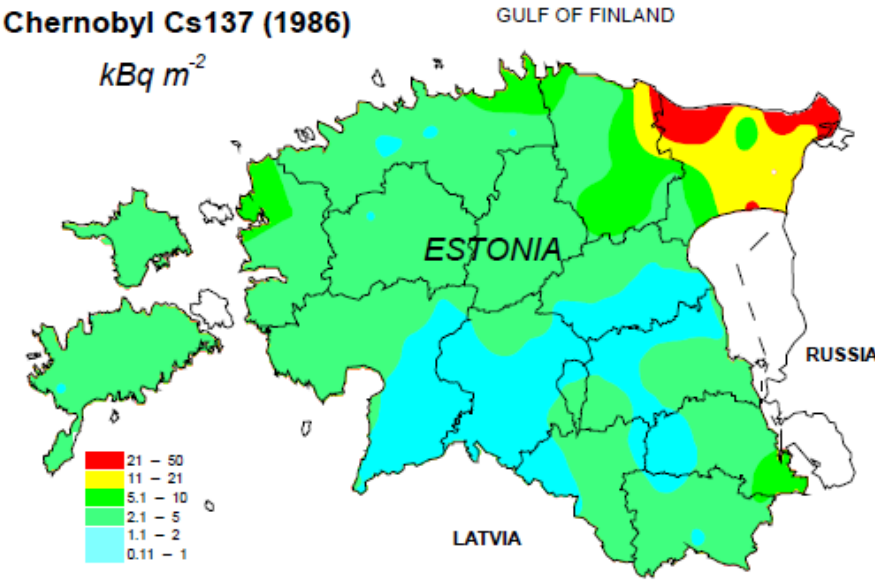

Figure 6. ^{137}Cs deposition caused by the 1986 Chernobyl accident in Estonia [53].

Radiocaesium is considered to be one of the most hazardous environmental contaminants due to both external gamma exposure and internal exposure via the human food chain. The areal distribution of fallout and the subsequent vertical distribution of radionuclides in the soil [54] has merited great attention. The latter is particularly important, as the transfer of radionuclides to man via terrestrial pathways (plant uptake, resuspension into the air, contamination of groundwater) depends on the vertical distribution of the radionuclides in soil.

The translocation of long-lived fallout radionuclides within soil strongly affects the time dependence of its entry into the human food supply via plant root uptake and, for gamma-emitting isotopes, also the dose rate due to external irradiation. Since the mobility of radionuclides in soil depends on complex physical, chemical and biological processes, a black-box approach based on a multi-compartmental model [55, 56] is frequently exploited for describing the radionuclide migration and accumulation in soils. Dose component estimation due to radiocaesium fallout in Estonia is given in Chapter 6.3.

4.7. Medical exposure

The announcement of Roentgen's discovery of X-rays in 1895 was almost immediately followed by medical use of X-rays. For over a century, the diagnostic use of X-rays has been routine. Radiation exposures from diagnostic medical examinations are generally low, and are almost always justified by the benefits of accurate diagnosis of possible disease conditions. Therapeutic uses of radiation naturally involve higher exposures and physicians will consider the risks of the treatment against the potential benefits [57]. Standardized radiation dose estimates can be given for a number of typical diagnostic medical procedures. Doses will change, depending on a number of variables, including the specific machine and manufacturer (in the case of radiology), study techniques (in the case of machine settings on the device used to produce the radiation, in nuclear medicine on the amount of activity administered and the patient's metabolism), and other factors [58].

In radiation therapy, much higher doses are given, with the intent of destroying cancer tissues. The intent is to provide a sufficient dose to kill unhealthy tissues while minimising damage to healthy tissues. This can be accomplished in a number of ways. With external radiation, many techniques are used to focus the radiation dose in the area of interest and give lower doses to normal tissues. Small sources may be placed very near to or in direct contact with cancer tissues (brachytherapy) and only left in place for defined periods of time. Therapy is also performed with internal radioactive sources, similar to those used in diagnostic nuclear medicine. These sources are attached to special molecules that are designed to be taken up preferentially by cancer cells, and less by other organs and tissues, thus leading to a more positive outcome without compromising the health of the patient. It is not possible to give specific dose calculations for these cases. Each situation must be carefully analysed by a radiation physicist before the therapy procedure. Doses for therapeutic procedures are typically in the order of hundreds or thousands times higher than in the diagnostic studies described above [59].

Table 8. Examples of the average doses of selected medical procedures [5, 60]

Procedure	**Effective Dose (mSv)**
Chest/ribs X-ray	0.02
Ankle X-ray	0.002
Lumbar spine X-ray	1.0
CT head	2.0
CT chest	11
Lithotripsy	1.3

5. EXPERIMENTAL TECHNIQUES

5.1. Gamma-spectrometry

Gamma rays are physically exactly similar to all other forms (e.g., X rays, visible light, infrared, radio) of electromagnetic radiation, except much higher photon energy and frequency, and shorter wavelength. An X-ray or gamma-ray photon is uncharged and creates no direct ionization or excitation of the material through which it passes. The detection of gamma rays is therefore critically dependent on causing the gamma-ray photon to undergo an interaction that transfers all or part of the photon energy to an electron in the absorbing material. Of the various ways gamma rays can interact in matter, only three interaction mechanisms have any real significance in gamma-ray spectroscopy – photoelectric absorption predominates for low-energy gamma rays (up to several hundred keV), pair production predominates for high-energy gamma rays (above 5–10 MeV), and Compton scattering is the most probable process over the range of energies between these extremes. The atomic number of the interaction medium has a strong influence on the relative probabilities of these three interactions [61].

Gamma-ray spectroscopy is the quantitative study of the energy spectra of gamma-ray sources, as the gamma-ray spectrometer also determines the energies of the gamma-ray photons emitted by the source. There are two commonly used types of detectors: inorganic scintillators and germanium semiconductor detectors [62]. In the current work, germanium semiconductor detectors were used.

For the measurements, a low-background ORTEC HPGe gamma spectrometer with 42% efficiency and 1.7 keV resolution (at 1.3 MeV) and a HPGe planar detector gamma spectrometer (MULTISPECTRUM, BSI, Latvia) were used. For the estimation of efficiencies, a set of high quality certified reference materials, e.g. IAEA-RG-SET (K, Th, U), with densities similar to the samples and calculations were used. The efficiency calibration of the gamma spectrometry systems was performed with the radionuclide specific efficiency method in order to avoid any uncertainty in gamma ray intensities, as well as the influence of coincidence summation and self-absorption effects of the emitting gamma photons.

For the measurement of the soil samples, the soil cores up to 25 cm were cut into 2–3 cm thick sections and dried at 105°C for 24 h. Both the wet and dry densities of the core sections were determined. For analysis the corresponding sections of usually two soil cores were mixed, homogenised in a mortar and put into metallic beakers. The true coincidence corrections for gamma lines, e.g., the 604 keV gamma line of ^{134}Cs , were evaluated using the GESPECOR2.0 software.

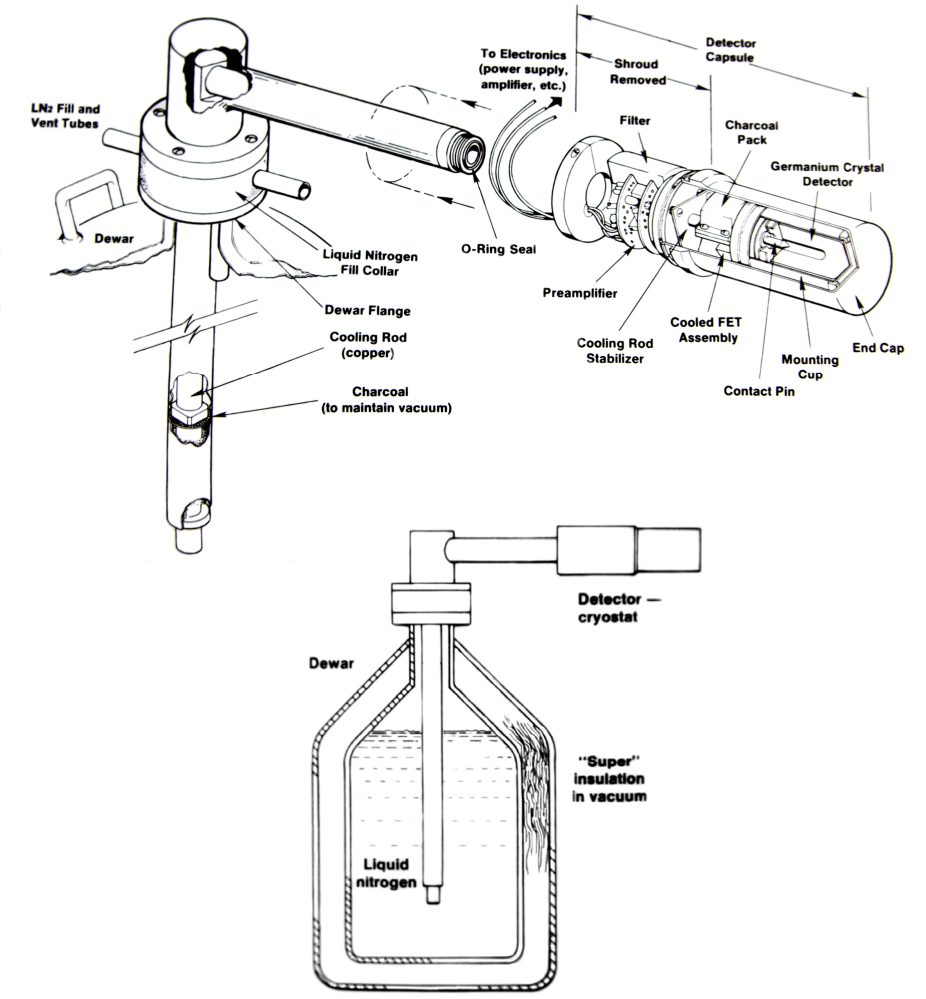

Figure 7. HPGe detector [61].

A hydraulic press was used to compress the Petrianov FPP airfilters to form a cylinder with the diameter of 41 mm, which were placed in the matching diameter beakers. ^{210}Pb content was analysed using the 46.5 keV line. As the sample heights varied, for each sample, the sample height, *h*, was measured and the attenuation coefficient, *m*, was determined using a ^{210}Pb source. Based on the measurement data of the reference samples of various heights, Monte Carlo modelling software (GESPECOR, GEANT4) was applied to develop a simple procedure for the activity correction on *m* and *h*.

All samples of building materials were crushed into grains, dried, homogenised and sealed into gas-tight metallic containers with a capacity of 120 cm^3. After closure radon and thoron could not escape, and during the ingrowth within about 3 weeks preceding the gamma spectrometric measurement a practical

equilibrium between ^{226}Ra and ^{214}Pb/Bi was achieved in the samples. For determination of the ^{226}Ra and ^{232}Th content in samples the photopeaks of their daughters ^{214}Pb, ^{214}Bi and ^{228}Ac, ^{208}Tl, respectively, were used.

5.2. Soil sampling

In order to achieve the most representative measurements, while selecting the sampling locations, the landscape and the specific geochemical make-up of the area were taken into account. Soil profiles were collected in undisturbed natural meadows. In addition, in order to eliminate the site-specific variability and to enable a better understanding of vertical migration features, a few samples were taken from the same sampling locations in the course of several years. Soil profiles down to a depth of ~ 20 (25) cm were collected by means of a 9 cm diameter stainless steel soil corer (SC). The design of the SC was based on an original idea that minimizes the compression of the soil during sample collection. This is extremely important to have correct and reproducible data for depth profile. In ordinary SC designs the compression is dependent on soil properties. In Fig. 8 the hardened sharp cutting edge (1) diameter of the SC is 2mm smaller from inner diameter of the rest tube (2). The optimal cutting edge angle is 15° that minimizes the compression soil length still leaving it enough sharp to cut it properly [63]. Also it facilitates the extraction of samples out from the SC. Samples are delicately pressed out from the other end of SC by using a special aid tool (3). The whole SC construction was based on a modular design that enables the simple replacement of any component in case of damage.

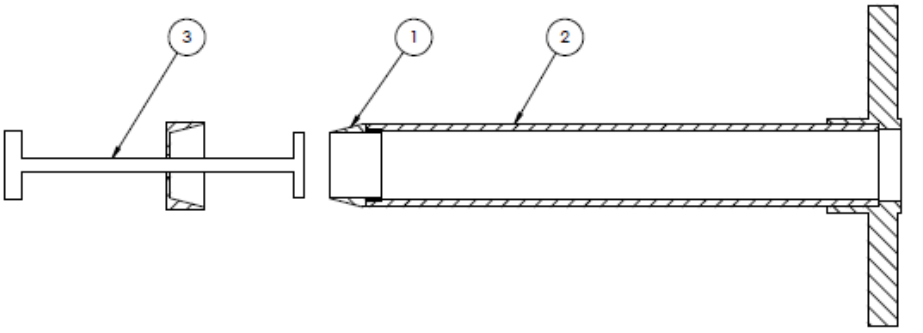

Figure 8. The 9 cm diameter stainless steel soil corer. (1) the hardened sharp cutting edge, (2) the rest tube and (3) special aid tool.

During the sampling process cylindrical samples were collected and wrapped in plastic film. This helped to transport the collected samples to the laboratory without any structural change. Typically two soil cores within 2–3 m were taken from each sampling location.

6. RESULTS AND DISCUSSION

6.1. Air filters (Paper I,VI,VIII, IX)

Relatively short-period data were available on the ^{210}Pb concentrations in surface air in Estonia [64]. Our aim was to observe ^{210}Pb concentration for longer time periods, and for this reason the archived aerosol filter samples collected weekly from 2001 to 2005 by the Estonian Radiation Protection Centre (currently Environmental Board) in the framework of the national radiation surveillance program were used for ^{210}Pb analysis. The filters were collected from the following sampling site: Harku-Tallinn meteorological station (59°23'54" N, 24°36'15" E), in the North of Estonia.

The following daily meteorological data were collected at the local automatic station: precipitation (rain, snow), relative humidity, air pressure, temperature and wind speed, were provided by the Estonian Meteorological and Hydrological Institute. The corresponding weekly and monthly averages needed for the statistical analysis of correlations of ^{210}Pb concentrations were calculated using meteorological parameters.

The ^{210}Pb activity concentration in the air demonstrated a considerable temporal variation (Fig. 9). The highest values occurred in the winter, especially during periods characterized by long-term high atmospheric pressure and low temperatures, i.e. during the governing of anticyclonic conditions. The most pronounced seasonal trend was that the winter months were characterized by the highest and the spring months by the lowest concentration values.

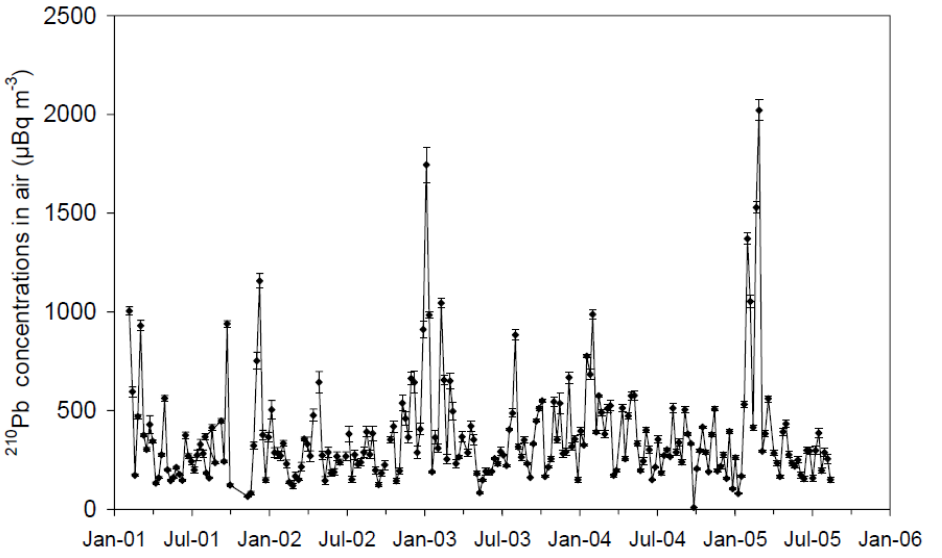

Figure 9. Weekly variation of 210 Pb activity concentrations in air, Harku-Tallinn, February 2001 – August 2005.

The low spring and summer concentrations are the result of an efficient vertical mixing generated by intense solar radiation in the troposphere and by large mixing layer heights. [65]. We made an attempt to search for correlations between measured activity concentration and the local meteorological parameters. Monthly-averaged ^{210}Pb activity concentrations exhibited significant correlations with air temperature: lower air temperatures facilitated higher ^{210}Pb activity concentrations. Another significant parameter was the average wind speed.

Based on UNSCEAR estimate [4] and assuming both ^{210}Pb and ^{210}Po, the annual dose due to this component is of the order of 0.09 mSv. Dose caused by ^{210}Pb in the air is determined mainly by the ingestion pathway. Doses caused by inhalation, external exposure from ^{210}Pb deposited in the ground and the immersion component are estimated (using MicroShield software) as negligible, 2 nSv/a, 3.6 nSv/a and 0.0002 nSv/a, respectively.

6.2. Building materials (Paper II)

Over the last 10 years, 53 samples of building materials were analyzed. The sample selection consisted primarily of commonly available materials which were obtained from building supply stores. The measurement results are presented in Tables 9 and 10. This data was used for the dose estimation.

Radionuclides presented in the most commonly used building materials are of the greatest interest. The biggest differences occur in the concentration of ^{40}K, where the lowest mean value is 145 Bq kg^{-1} for Misso Light brick and highest 449 Bq kg^{-1} for Aseri III brick. Much smaller variations are found for other radionuclides. From the results it can be seen that the lowest mean value of ^{226}Ra concentration is 12.6 Bq kg^{-1} measured in Misso Light clay brick, while the highest mean value for the same radionuclide is 27.3 Bq kg^{-1} for oilshale ash block.

Table 9. Mean activity concentrations of radionuclides (Bq kg^{-1}) in the most commonly used building materials in Estonia.

Material	**^{226}Ra**	**^{238}U**	**^{232}Th**	**^{40}K**
Aseri I brick	21.1±3.0	33.9±4.3	29.6±3.2	436±20
Aseri II brick	20.3±2.9	32.6±4.2	29.7±3.1	436±20
Aseri III brick	19.6±2.8	31.5±4.0	30.9±3.1	449±21
Misso Light brick	12.6±1.5	19.6±2.0	13.2±1.4	145±7
Misso Dark brick	18.4±2.4	28.8±3.2	19.4±2.2	207±9
Ash-blocks	27.3±2.3	28.5±2.2	14.1±1.7	308±8

Table 10. Mean activity concentrations of radionuclides (Bq kg^{-1}) in some building materials used in Estonia

Material	**^{226}Ra**	**^{238}U**	**^{232}Th**	**^{40}K**
Finnish building clay	53.6±3.4	56.3±3.3	58.5±3.2	748±17
Siimusti building clay	69.3±3.8	70.6±3.4	82.0±3.5	678±15
German building clay	35.6±2.3	36.9±2.1	55.2±2.4	447±11
Gypsum board	4.4±1.0	4.9±1.1	0.8±0.2	7.0±0.4
Grosso 311 floor tiles	64.1±3.8	48.4±2.4	81.5±2.8	344±9
Pronto 147 floor tiles	48.7±2.5	50.3±2.2	86.3±2.8	284±7
Kunda cement	46.9±2.8	49.0±2.5	21.4±1.6	587±13
Kolumbia stone	25.5±1.8	26.3±1.7	28.4±1.6	480±11
Concrete	35.1±2.1	36.1±1.8	11.3±0.9	207±5

Table 11. Index *I* and radium equivalent values calculated for the measured building materials.

Material	**Index *I***	**Radium equivalent *Ra(eq)***
Aseri I brick	0.47	97
Aseri II brick	0.44	96
Aseri III brick	0.37	98
Misso Light brick	0.16	66
Misso Dark brick	0.23	62
Ash-blocks	0.26	64
Finnish building clay	0.72	196
Siimusti building clay	0.87	239
German building clay	0.54	149
Gypsum board	0.02	6
Grosso 311 floor tiles	0.74	207
Pronto 147 floor tiles	0.69	194
Kunda cement	0.46	123
Kolumbia stone	0.39	103
Concrete	0.24	67

The calculations of annual doses from exposures by naturally occuring radionuclides present in building materials of dwellings were performed using MathCAD software. The method used for estimation of the doses was based on the work by Mustonen, [66] and Markkanen [67]. In these calculations, a rather conservative assumption that all construction elements of the room, e.g., walls, the ceiling and the floor, are made of the same material is accepted. As a result, the evaluated annual doses represent the maximum values for the corresponding

building material. The evaluations are not made for the materials, e.g., the binding materials and cover tiles, which practically form a relatively small mass fraction of the construction element. The density of building materials used in calculations is taken equal to 2350 kg m^{-3}. In dose calculations the attenuated fractions of 80% of cosmic radiation [3] and 5–10% of the outdoor background terrestrial radiation, estimated by MicroShield modeling, are not taken into account.

The clay bricks are widely used for dwelling construction, and their natural radionuclide content provides radiation exposure to the population. As there is concern that some of the buildings will cause excessive radiation doses to the residents due to gamma rays emitted, the data obtained from our measurements was used for the estimation of corresponding doses to the population. Based on the results it can be concluded that in dwellings constructed from those building materials measured in the course of the present study, the total effective dose rate is less than 1 mSv per year.

Tabel 12. Total annual dose due to natural radionuclides in Estonian building materials (mSv).

Material	**Total annual dose (mSv)**
Aseri I brick	0.42
Aseri II brick	0.42
Aseri III brick	0.44
Misso Light brick	0.16
Misso Dark brick	0.23
Ash-blocks	0.25
Cement	0.30

6.3. Radiocaesium (Paper III, IV,V,VII)

In order to study the vertical migration of radiocaesium in soils, the depth distributions of the ^{134}Cs and ^{137}Cs activity concentration have been measured for undisturbed soil profile samples collected at more than 30 locations in NE Estonia since 1991 (see Fig. 10).

Some typical results of the averaged depth distributions of the measured ^{134}Cs and ^{137}Cs activity concentrations in a 1 m^2 soil layer of 1 cm thickness in the profile sections (% m^{-2} cm^{-1}) collected in NE Estonia between 1991 and 2003 are presented in Fig. 11. Activity concentrations are presented as the percentages of activity of the profile to the total at the collection time.

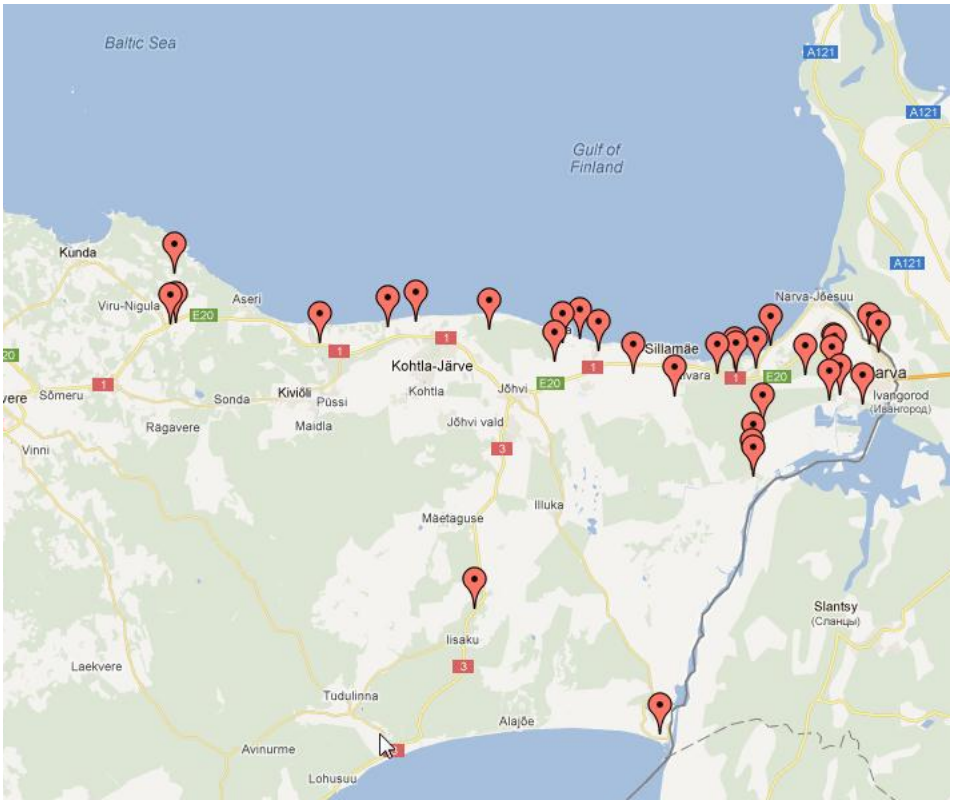

Figure 10. Map of North Eastern Estonia showing sampling sites (GoogleMap).

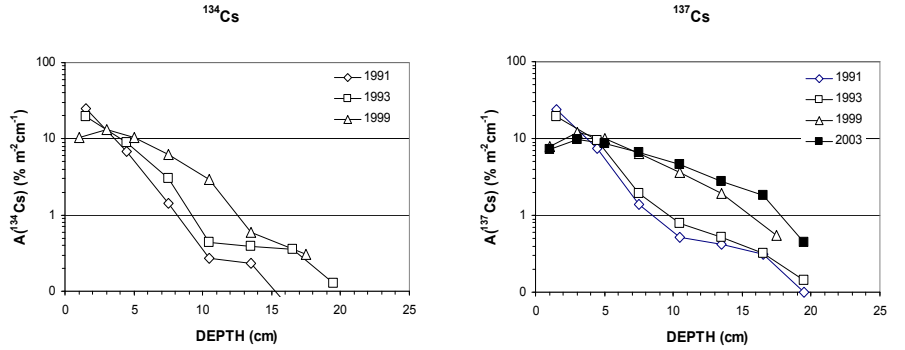

Figure 11. Averaged depth-dependences of ^{134}Cs and ^{137}Cs activity concentrations (% m^{-2} cm^{-1}) in soil profiles.

Considering the radionuclide transport in soil as a 1D diffusion-convection process has led to multiple models and their modifications, resulting in the depth-distributions of the Gaussian, the Lorentzian or the lognormal types [68]. To describe the depth-dependence of the ^{137}Cs concentration arising from long-term transport we applied the lognormal distribution of the following form:

$$C(x) = \frac{A}{\sqrt{2\pi}\, wx} \exp\left[-\frac{(\ln x - \ln xc)^2}{2w^2} \right],$$

where x is the depth, while its parameters are the mean, *ln xc,* and the variance of the distribution, w^2. According to theory [69], these parameters are time-dependent functions of the diffusion coefficient and the mass flow transport velocity, respectively.

For use of the compartmental modeling the soil was split into 4 horizontal layers which were connected by downward/ upward transport rates of the radionuclide under study. The horizontal layers (compartments) were chosen at the following depths: 0–1 cm (*a*), 1–5 cm (*b*), 5–15 cm (*c*) and 15–30 cm (*d*). This selection of uniformly mixed compartments was considered a reasonable compromise for an adequate description of migration and uptake processes of radionuclides in soil [70]. The top 1 cm of soil accepts the deposited radionuclides, participates in resuspension, etc. Radionuclides in the lower compartments extending to 15 cm in depth are considered available for root uptake by different plants. The compartmental model we used to describe the time dependent migration of radiocaesium in soil was the following:

$$\frac{dA}{dt} = \lambda_{ba} B - \lambda_{ab} A - \lambda_0 A$$

$$\frac{dB}{dt} = \lambda_{ab} A - \lambda_{ba} B - \lambda_{bc} B - \lambda_0 B$$

$$\frac{dC}{dt} = \lambda_{bc} B - \lambda_{cd} C - \lambda_0 C$$

$$\frac{dD}{dt} = \lambda_{cd} C - \lambda_{dv} D - \lambda_0 D$$

where

A, B, C and *D* are the activities of ^{134}Cs or ^{137}Cs in the corresponding compartments *a, b, c, d* (in Bq per 1 m^2) at time *t*,

λ_{ab}, λ_{bc}, λ_{cd}, λ_{ba} are the transfer rates (y^{-1}) between compartments, respectively,

λ_{dv} is the transfer rate (y^{-1}) from compartment *d* to the sink *v*,

λ_0 is the radioactive decay constant (y^{-1}), which equals 0.3357 ± 0.0002 y^{-1} for ^{134}Cs and 0.02310 ± 0.00002 y^{-1} for ^{137}Cs.

During the first ten years after deposition, the ^{134}Cs activity is mostly retained in the compartments *a* and *b,* i.e., at the depth down to 5 cm, while ^{137}Cs is distributed down to 15 cm (compartments *a, b* and *c*). The activities in the lowest compartment *d* and the outflow to the sink *v* were rather low. In this way, ^{137}Cs stays available for soil-plant transfer and results in the internal exposure of the human population over a considerable time following the deposition (10–20 years). The observed weak time-dependences of the ^{137}Cs depth-distribution parameters demonstrated a rather slow migration rate of radiocaesium to deeper soil layers and its strong retention in the near-surface soil. This means that there was broadening of the distribution and a shift of its maximum concentrations to deeper soil layers in time.

Using the modelled time-dependent activities of ^{134}Cs and ^{137}Cs in the depth profile the total depositions 10 kBq m^{-2} for ^{137}Cs and 5 kBq m^{-2} for ^{134}Cs, the effective dose rate (nSv h^{-1}) and its time-dependence due to the migration and the radioactive decay of radionuclides were estimated. Based on these results and assuming outdoor exposure during 1760 h y^{-1}, the annual effective dose from this exposure was also evaluated (Fig. 12). Immediately after the deposition, the effective dose rate caused by gamma radiation of ^{134}Cs dominated, while after a year the ^{137}Cs component prevailed. In different locations of NE Estonia the dose rate values varied in the range from 14 nSv h^{-1} to 177 nSv h^{-1}. The mean external dose rate for the region was 80 nSv h^{-1}. These short-time dose rates, which were comparable to those caused by the local natural background radiation, decreased rapidly over time, due to radioactive decay and the migration of radiocaesium to deeper soil layers. The effective half-time of dose rate decrease showed significant increase over time, being 2.2 y during the first year, 4.8 y from 2 to 10 y and about 10.4 y after 10 y from the deposition. Considering the found total depositions of radiocaesium in soil from the Chernobyl 1986 accident, the 50 year effective doses caused by external gamma exposure varied in the range from 0.13 mSv to 1.74 mSv, with the mean of 0.79 mSv in NE Estonia. The calculated dose and dose rate values should be considered as the conservative references, because in their derivation the uncertainties caused by the observed significant variations in areal and depth distribution of radiocaesium, in soil composition, water content and density, etc., were not evaluated.

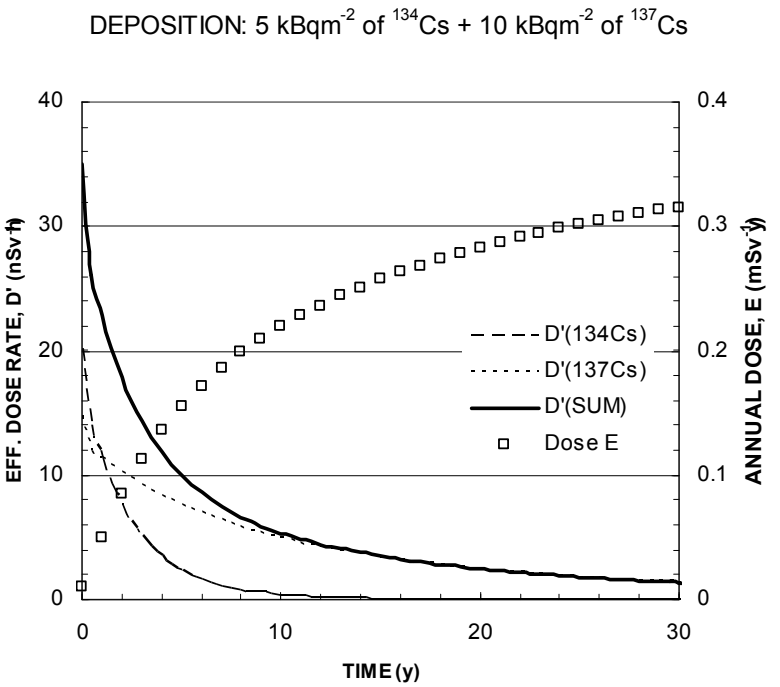

Figure 12. Time-dependence of effective dose rate of external exposure, D'(nSv h^{-1}) and annual effective dose, E (mSv y^{-1}), at 1 m above soil, assuming deposition of 5 kBq m^{-2} of ^{134}Cs and of 10 kBq m^{-2} of ^{137}Cs.

6.4. Estimation of the annual dose to Estonian population

Taking into account the different components, the annual dose to Estonian population can be estimated. This dose takes into account only the components caused by existing situations. Additionally, there are planned exposure and medical exposure situations. There is information about occupational doses available in Estonia, but there is limited information available about the doses due to medical exposures.

Table 13. Annual dose for Estonian population (mSv) caused by existing exposures.

	Outdoor	**Indoor**	**Ingestion**	**Sum**
Radon	–	2.1	–	2.1
Terrestrial gamma radiation	0.10 (0.02 … 0.41)	0.02 (0.01 … 0.10)	–	0.12 (0.03 … 0.52)
Cosmic radiation	0.07	0.24	–	0.31
Foodstuff	–	–	0.2	0.2
Drinking water	–	–	0.14 [(1)]	0.14
Electricity production	0.001	–	0.0006	0.0016
^{210}Pb	–	–	0.09	0.09
Building materials	–	0.27 (0.16 ... 0.44)	–	0.27 (0.16 … 0.44)
Radiocaesium[(2,3)]	0.0006 (0 … 0.015)	0	0.0003 (0… 0.008)	0.0009 (0 … 0.023)
TOTAL	**0.17**	**2.63**	**0.43**	**3.23**

[(1)]Assuming that ca 1 million people use water from non Cm-V WTZ
[(2)] In the estimation of the doses an estimate of "external/internal" ratio of ca 2 was used [4].
[(3)] Assuming country-wide average deposition of the Chernobyl ^{137}Cs to be 2 kBq m^{-2} and using the migration model described above.

The estimated annual dose 3.23 mSv is a bit higher than the average dose due to natural sources of radiation estimated by UNSCEAR – 2.4 mSv [4]. The main difference is caused by the dose caused from radon.

7. CONCLUSIONS

The current studies provided the ability to estimate several missing components which were needed in order to properly estimate the overally annual radiation exposure levels for the Estonian population due to exisiting exposures.

1. Estimated weekly activity concentrations of ^{210}Pb in aerosol filter samples varied from 0.065 mBq m^{-3} to 2.02 mBq m^{-3} with arithmetic mean value of 0.37 mBq m^{-3} and followed a long-normal distribution with a geometric mean value of 0.31 mBq m^{-3} and dispersion factor of 0.52 in Tallinn-Harku. In Narva-Jõesuu and in Tõravere the corresponding arithmetic mean values of 0.45 mBq m^{-3}and 0.51 mBq m^{-3} are found, so the country-wide average is 0.44 mBq m^{-3}.
2. ^{210}Pb in the air causes annual dose component of the order of 0.09 mSv/a, it is determined primarily via the ingestion pathway.
3. Based on the study of the building materials it was concluded that all the materials passed the accepted regulatory standards and may be used without restrictions. The mean values of the index *I* of the studied building material samples range from 0.02 to 0.74.
4. The average effective dose annually received by the Estonian residents due to the building materials vary from 0.16 mSv to 0.44 mSv. Only a few of the doses are marginally larger than the world-wide average annual external effective dose of 0.41 mSv from natural indoor radiation sources [3].
5. Compartmental modeling described the time-dependent migration and allowed the determination of radiocaesium transfer rates in soil compartments and to predict the vertical distribution of both ^{134}Cs and ^{137}Cs in soil at different times following deposition.
6. Considering the found total deposition levels of radiocaesium in the soil as a result of the 1986 Chernobyl accident, the 50 year effective dose caused by external gamma exposure varied in the range from 0.13 mSv to 1.74 mSv, with the mean of 0.79 mSv in NE Estonia.
7. Overall, the annual radiation dose of Estonian population from existing exposures was estimated to be 3.23 mSv.

SUMMARY

For assessment of the dose to Estonian population there is a need to assess the doses at least from the all major components. During the years several components have been assessed; however there was still lack of some important components, which were taken as aim of this study. In the current study the following components were assessed – ^{210}Pb concentration in the air; natural radionuclides in the building materials and radiocaesium from nuclear weapons testing and Chernobyl accident fallout. The activity concentrations in the samples were determined by gamma ray spectrometry.

^{210}Pb content was measured using aerosol airfilter samples collected weekly from Harku during the period of 2001–2005. Activity concentrations in air of Harku varied from 0.065 mBq m^{-3} to 2.020 mBq m^{-3} with an arithmetic mean value of 0.37 mBq m^{-3} and followed a long-normal distribution with a geometric mean value of 0.31 μBq m^{-3} and dispersion factor of 0.52. The mean values were slightly higher than those expected at a location of comparable latitude and longitude. The analysis of monthly averaged concentrations revealed a dominant seasonal variation governed mainly by the origin of intruding air masses: high ^{210}Pb concentrations in winter and low values in spring and summer. The annual dose due to this component is of the order of 0.09 mSv. Dose caused by ^{210}Pb in the air is determined mainly by the ingestion pathway.

Radionuclides naturally occurring in building materials may significantly contribute to the annual doses. As information on the radioactivity of such materials is lacking, the study of building materials used in Estonia was carried out in order to estimate the annual dose to the Estonian population due to natural radionuclides in building materials. During the study 53 samples of commonly used raw materials and building products were collected and measured. Their mean values were in the ranges 7–747 Bq kg^{-1} for ^{40}K, 4.4–69 Bq kg^{-1} for ^{226}Ra, and 0.8–86 Bq kg^{-1} for ^{232}Th. The activity index I in the 53 different building materials varied from 0.02 to 0.74 and the radium equivalent, from 6 to 239. The average annual dose for the people, caused by the building materials of dwellings, was assessed for most commonly used materials. It was estimated to be in the range from 0.16 mSv to 0.44 mSv.

The deposition and vertical depth distribution of ^{134}Cs and ^{137}Cs in the natural undisturbed soil profiles down to 20–25 cm were studied at locations in the North Eastern Estonia, which were most strongly affected by the Chernobyl fallout in 1986. The total depositions were estimated based on summing the sampled and measured activities of ^{134}Cs and ^{137}Cs in all sections of soil profiles. The Chernobyl ^{137}Cs deposition values varied considerably from site to site and the range was from 3.9 kBq m^{-2} to 50.2 kBq m^{-2}, with the average of 22.8 kBq m^{-2} for the region (reference data May 1, 1986). The ratio of total activities, A(^{134}Cs)/A(^{137}Cs), varied in the range from 0.47 to 0.55.

Using a compartmental model and the observed data on the ^{134}Cs and ^{137}Cs activity concentrations (Bq m^{-2}) in four compartments, 0–1 cm, 1–5 cm, 5–15 cm and 15–30 cm, of soil collected in 1991–2003, the approximate residence half-times of radiocaesium in soil were determined. The latter increased from 3.7 y in the top-most compartment to 8.6 y and 36 y in the next compartments, respectively.

The time dependence of the external gamma-dose rate at the height of 1m above a flat ground area arising from the deposited and migrating radiocaesium was calculated using the modeled data on the ^{134}Cs and ^{137}Cs activity in soil compartments. Considering the total depositions of radiocaesium in soil from the Chernobyl 1986 accident, the 50 year effective doses caused by external gamma exposure varied in the range from 0.13 mSv to 1.74 mSv, with the mean of 0.79 mSv in the region.

Taking into account the previous studies about the components of the dose to Estonian population and adding the results of the current study, we can conclude that the average annual dose to member of the public is 3.23 mSv. There is only one major component missing from this assessment and this is doses due to the medical exposures.

SUMMARY IN ESTONIAN

Elanikkonna kiirgusdoosi komponentide hindamine

Elanikkonna kiirgusdoosi hindamiseks Eestis on vaja hinnata seda moodustavad komponendid. Mitmed nendest on erinevate uuringute raames juba hinnatud, kuid seni puudusid veel paari olulise komponendi poolt põhjustatava elanikudoosi hinnangud. Käesolevas töös vaadeldi eelkõige ^{210}Pb sisaldust õhus, looduslike radionukliide ehitusmaterjalides ning nii atmosfääris läbiviidud tuumakatsetuste kui ka Tšernobõli avarii tulemusena sadenendud radiotseesiumit. Proovide analüüsiks ning radionukliidide kontsentratsioonide määramiseks kasutati kõrge eraldusvõimega HPGe gammaspektromeetrilist analüüsimeetodit.

^{210}Pb kontsentratsioonide määramiseks õhus kasutati Harku filterjaamadest kogutud filtreid. Filtreid eksponeeriti nädala aja jooksul ning proovid pärinevad perioodist 2001–2005. Eriaktiivsused Harkus varieerusid vahemikus 0.065 mBq m^{-3} kuni 2.02 mBq m^{-3}, aritmeetiline keskmine 0.37 mBq m^{-3}. Keskmised väärtused on natuke kõrgemad kui võrrelda sarnastel pikkus- või laiuskraadidel mõõdetud tulemustega. Kasutades eriaktiivsuste kuiseid keskmistatud väärtusi, siis võis järeldada, et oluliseks on sissetulevad õhumassid – kõrgemad eriaktiivsuste väärtused on talvistel perioodidel ning madalaimad kevadel ja suvel. Aastane elaniku kiirgusdoos sellest komponendist tulenevalt on suurusjärgus 0.09 mSv ning see on eelkõige tingitud sisedoosist.

Ehitusmaterjalides leiduvad looduslikud radionukliidid võivad olla väga olulisteks elanikudoosi tekitajateks. Käesoleva uurimistöö raames määrati looduslike radionukliidide sisaldus 53 erinevas Eestis kasutatava ehitusmaterjali proovis. Leiti, et looduslike radionukliidide ^{40}K, ^{226}Ra ja ^{232}Th aktiivsuse kontsentratsioonid varieeruvad uuritud ehitusmaterjalides järgmistes vahemikes: vastavalt 7–747 Bq kg^{-1}, 4.4–69 Bq kg^{-1} ning 0.8–86 Bq kg^{-1}. Aktiivsuse kontsentratsioonide alusel hinnatud ehitusmaterjalide aktiivsusindeksi I väärtused asuvad piirides 0.02 kuni 0.74. See viitab sellele, et ehitusmaterjalide kasutamisel ei ole vaja kasutada piiranguid. Levinumate ehitusmaterjalide jaoks tehti doosihinnangud siseruumides ja selle alusel saadud elaniku aastased kiirgusdoosid jäävad vahemikku 0.16–0.44 mSv.

Radiotseesiumi (^{134}Cs ja ^{137}Cs) käitumise uurimiseks kasutati 20–25 cm sügavusi pinnaseproove Kirde-Eestis. Proovid võeti sellest piirkonnast eelkõige seetõttu, et Tšernobõli avarii tulemusena tuvastati seal suurimad sadenemised maapinnale. Pinnaseproove võeti enam kui 30 punktis ning nendes proovivõtu kohtades varieerusid radiotseesiumi hinnatud sadenemised vahemikus 3.9 kBq m^{-2} kuni 50.2 kBq m^{-2}. Töö käigus tuvastati, et kasutades neljast osast koosnevat plokkmudelit oli võimalik kirjeldada radiotseesiumi käitumist pinnases ning samuti võimaldab see määrata rresidentsi poolestusaega.

Kasutades modelleerimise andmeid tuvastati pinnases leiduva radiotseesiumi poolt põhjustatavad doosikiirused 1 meetri kõrgusel maapinnast. Võttes arvesse

Tsernobõli avarii tulemusena toimunud sadenemised, siis hinnanguliselt põhjustab see 50 aastase efektiivdoosi vahemikus 0.13 kuni 1.74 mSv (keskmine 0.79 mSv).

Võttes arvesse juba teadaolevad elanikudoosi komponendid ning lisades siis juurde käesoleva töö raames hinnatud, võib väita, et Eesti elaniku kiirgusdoos on keskmiselt 3.23 mSv aastas. Sellele lisandub veel meditsiinist põhjustatav doos.

ACKNOWLEDGEMENTS

This work is dedicated to Küllike Realo.

I would like to express my greatest gratitude to my supervisors Enn Realo and Rünno Lõhmus for their guidance and support. And I am so greatful to everybody who knowingly or unknowingly introduced me ionizing radiation and the system of radiation protection. Thank to you the life has provided so many interesting challenges!

This work has been partially supported by graduate school „Functional materials and technologies" receiving funding from the European Social Fund under project 1.2.0401.09-0079 in Estonia.

REFERENCES

[1] ICRP 103, 2007. The 2007 Recommendations of the International Commission on Radiological Protection ICRP Publication 103; Ann. ICRP 37.

[2] IAEA, 2004. Radiation, People and the Environment, Editor J. Ford.

[3] UNSCEAR, 2000. Report on Sources and Effects of Ionizing Radiation to the General Assembly (Annex B) Vienna.

[4] UNSCEAR, 2010. United Nations Scientific Committee on the Effects of Atomic Radiation. Sources and Effects of Ionizing Radiation. 2008 Report to the General Assembly, with scientific annexes. United Nations, New York.

[5] Watson, S.J., Jones, A.L., Oatway, W.B., Hughes, J.S., 2005. Ionising Radiation Exposure of the UK Population 2005 Review, HPA-RPD-001.

[6] NCRP, 2009. Report No. 160 – Ionizing Radiation Exposure of the Population of the United States.

[7] Pahapill, L., Rulkov, A., Rajamäe, R., Akerblom, G., 2003. Radon in Estonian dwellings. Results from a National Radon Survey, SSI:2003:16.

[8] Hall, E., 2006. Radiobiology for the radiologist. Lippincott Williams & Wilkins.

[9] ICRP 101, 2006. Assessing Dose of the Representative Person for the Purpose of Radiation Protection of the Public and The Optimisation of Radiological Protection: Broadening the Process. Volume 36 No 3.

[10] IAEA, 2011. Radiation Protection and Safety of Radiation Sources: International Basic Safety Standards.

[11] Estonian Radiation Act, 2004.

[12] Estonian Radiation Protection Centre, 2008. Presentations of the seminar.

[13] WHO, 2009. WHO Handbook on Indoor Radon. A Public Health Perspective.

[14] EC, 2005. An Overview of Radon Surveys in Europe, G. Dubois.

[15] Billon, S., Morin, A., Caer, S., Baysson, H., Gambard, J.P., Backe, J.C., Rannou, A., Tirmarche, M., Laurier, D., 2005. French population exposure to radon, terrestrial gamma and cosmic rays. Radiat Prot Dosimetry, 113:314–320.

[16] Menzler, S., Piller, G., Gruson, M., Rosario, A. S., Wichrnann, H. E., Kreienbrock, L., 2008. Population attributable fraction for lung cancer due to residential radon in Switzerland and Germany. Health Phys, 95:179–189.

[17] WHO, 2007. International Radon Project Survey on Radon Guidelines, Programmes and Activities.

[18] Darby S., Hill D., Auvinen A., Barros-Dios J. M., Baysson H., Bochicchio F., Deo H., Falk R., Forastiere F., Hakama M., Heid I., Kreienbrock L., Kreuzer M., Lagarde F., Mäkeläinen I., Muirhead C., Oberaigner W., Pershagen G., Ruano-Ravina A., Ruosteenoja E., Schaffrath Rosario A., Tirmarche M., Tomásek L., Whitley E., Wichmann H.-E. and Doll R., 2005. Radon in homes and risk of lung cancer: collaborative analysis of individual data from 13 European case-control studies. BMJ; 330(7485):223-.

[19] Lepasson, M., Tänavsuu, K., Lust, M., 2008. Indoor radon: problems and surveys in Estonia. Proceedings of International Conference on Radioecology and Environmental Radioactivity, Bergen 2008.

[20] Pahapill, L., Rulkov, A., 2004. Radon in dwellings, project report of environment management programme. Tallinn.

[21] Petersell, V., Akerblom, G., Eek, B.-M., Enel, M., Mõttus, V., Täht, K., 2005. Radon Risk Map of Estonia. Tallinn-Stockholm.

[22] Realo, E., 1998. Dose-Rates in the Estonian Environment. In J. Søgaard-Hansen and A. Damkjær (Eds.) Proc. of the Regional IRPA Congress, June 12–13, 1998, Stockholm. Risoe National Laboratory, Roskilde, 1998, pp. 193–196.
[23] Holm, E., 1994. Radioecology. World Scientific, Singapore-New Jersey-London-Hong Kong.
[24] Winkler, R., Rosner, G., 2006. Seasonal and long-term variation of 210Pb concentration in air, atmospheric deposition rate and total deposition velocity in south Germant. Sci. Total Environ. 263, 57–68.
[25] Realo, K., Isakar, K., Lust, M., Realo, E. (2007). Weekly variations of the 210Pb air concentration in North Estonia. Boreal Environment Research, 12(1), 37–41.
[26] Beks, J.P., Eisma, U. D., van der Plicht, J., 1998. A record of atmospheric ^{210}Pb deposition in The Netherlands. Sci. Tot. Environ., 222:35–44.
[27] Tokieda, T., Yamanaka, K., Harada, K., Tsunogai, S., 1996. Seasonal variations of residence time and upper atmospheric contribution of aerosols studied with Pb-210, Bi-210, Po-210 and Be-7. Tellus, 48B: 690–702.
[28] Environmental Board, 2011. The Results of the Estonian radiation monitoring program in 2010
http://www.kiirguskeskus.ee/image/Kiirgusseire_aruanne_2010.pdf.
[29] Forte, M., Bagnato, L., Caldognetto, E., Risica, S., Trotti, F., Rusconi, R., 2010. Radium isotopes in Estonian groundwater: measurements, analytical correlations, population dose and a proposal for a monitoring strategy. Journal of Radiological Protection, 30(4): 761–780.
[30] Savitskaja, L., Savva, V., Jaštšuk, S., 2003. Estimation of the radiunuclides in the drinking water. K-11-1-2003/681 (in Estonian).
[31] Estonian Radiation Protection Centre, 2005. Assessment of Health Risks from consumtion of the drinking water (in Estonian).
[32] Parami, V. K., Sahoo, S. K., Yonehara, H. Takeda, S., Quirit, L. L., 2010. Accurate determination of naturally occuring radionuclides in Philippine coal-fired thermal power plants using inductively coupled plasma mass spectrometry and γ-spectroscopy. Microchemical Journal 95, 181–185.
[33] Realo, E., Realo, K., Jogi, J., 1996. Releases of natural radionuclides from oil-shale-fired power plants in Estonia. Journal of Environmental Radioactivity, 33(1), 77–89.
[34] Realo, E.; Realo, K. (2003). Natural radionuclides in surface soils in RA-RICH areas, NE Estonia. Abstracts of Papers of the American Chemical Society, 226, U81–U81.
[35] Lust, M., 1999. Estimation of the doses from energy production. MSc Thesis, University of Tartu.
[36] UNSCEAR, United Nations Scientific Committee on the Effects of Atomic Radiation, 1977. Sources, Effects and Risks of Ionizing Radiation. Report to the General Assembly with Annex B: Natural Sources of Radiation. United Nations, New York.
[37] UNSCEAR, 1993. United Nations Scientific Committee on the Effects of Atomic Radiation. Sources and Effects of Ionizing Radiation. United Nations, New York.
[38] Chen, C.J., Lin, Y.M., 1996. Assessment of building materials for compliance with regulations of ROC. Environmental International 22, S221–S226.
[39] Stoulos, S., Manolopoulou, M., Papastefanou, C., 2003. Assessment of natural radiation exposure and radon exhalation from building materials in Greece, Journal of Environmental Radioactivity 69, 225–240.

[40] Righi, S., Bruzzi, L., 2006. Natural radioactivity and radon exhalation in building materials used in Italian dwellings, Journal of Environmental Radioactivity 88, 158 – 170.
[41] Ahmed, N. K., 2005. Measurement of natural radioactivity in building materials in Qena city, Upper Egypt, Journal of Environmental Radioactivity 83, 91 – 99.
[42] Brıgido Flores, O., Montalvan Estrada, A., Rosa Suarez, R., Tomas Zerquera, J., Hernandez Perez, A., 2008. Natural radionuclide content in building materials and gamma dose rate in dwellings in Cuba. Journal of Environmental Radioactivity 99, 1834–1837.
[43] OECD-NEA., 1979. Organization for Economic Co-operation and Development—Nuclear Energy Agency. Report Exposure to Radiation from Natural Radioactivity in Building Materials. Paris.
[44] European Commission (EC), 1999. Radiation Protection 112. Radiological Protection Principles concerning the Natural Radioactivity of Building Materials. Directorate-General, Environment, Nuclear Safety and Civil Protection.
[45] El-Hussein, A., 2005. A study on natural radiation exposure in different realistic living rooms, Journal of Environmental Radioactivity 79, 355–367.
[46] Trevesi, R., Risica, S., D'Alessandro, M., Paradiso, D., Nuccetelli, C., 2012. Natural radioactivity in building materials in the European Union: a database and an estimate of radiobiological significane. Journal of Envrionmental Radioactivit 105, 11–20.
[47] Risica, S., Trevisi, R., Nuccetelli, C., 2012. Building material with enhanced or elevated levels of natural radioactivity: analyses of the use of index criteria for limiting their use. EU-NORM 1 Symposium Proceedings, 5–8 June 2012, Tallinn, Estonia.
[48] Webb, J., 1949. The fogging of photographic film by radioactive contaminants in cardboard packing materials. Phys. Rev. 76, 375–380.
[49] Palsson, S. E., 2012. Prediction of global fallout and assoicated environmental radioactivity. PhD Disserattion, University of Iceland.
[50] Eisenbund, M., Gesell, T.F., 1997. Environmental radioactivity: from natural, industrial and militar sources. 4th ed. Academic Press, San Diego.
[51] CTBTO, 2012. Locations of Nuclear Explosions, Interactiv map. http://www.ctbto.org/map/
[52] He, Q., Walling, D. E., 1997. The Distribution of Fallout ^{137}Cs and ^{210}Pb in Undisturbed and Cultivated Soils. Appl. Radiat. Isot. Vol. 48, No. 5, pp. 677–690.
[53] Realo, E., Jogi, J., Koch, R., Realo, K., 1995. Studies on radiocesium in Estonian soils. Journal of Environmental Radioactivity, 29(2), 111–119.
[54] Hölgye, Z., Maly, M., 2000. Sources, vertical distribution, and migration rates of $^{239, 240}$Pu, ^{238} Pu, and ^{137}Cs in grassland soil in three localities of central Bohemia. Journal of Environmental Radioactivity 47, 135–147.
[55] Frissel, M. J., Jakubik, A. T., van der Klugt, N., Pennders, R., Poelstra, P. & Zwemmer, E., 1981. Modeling of the Transport and Accumulation of Strontium, Cesium and Plutonium. Experimental Verification. Report 185-76-1 BIA N, Commission of the European Communities, Brussels, Belgium.
[56] Rühm, W., Kammerer, L., Hiersche, L. and Wirth, E., 1996. Migration of ^{137}Cs and ^{134}Cs in different forest soil layers. J. Environ. Radioactivity 33, 63–75.
[57] Martin, C. J., Sutton, D. G., 2002. Practical Radiation Protection in Healthcare. Oxford University Press.

[58] Cherry, S. R., Sorenson, J. A., Phelps, M. E., 2003. Physics in Nuclear Medicine. Saunders.
[59] Statkiewicz Sherer, M. A., Visconti, P. J., Ritenour, E. R., 2002. Radiation Protection in Medical Radiography, Mosby.
[60] Hart, D., Wall, B. F., 2002 Radiation Exposure of the UK Population from Medical and Dental X-ray Examinations. Chilton, NRPB-W4.
[61] Knoll, G. F.,1989. Radiation Detection and Measurement. Second Edition. John Wiley & Sons
[62] Gilmore, G., Hemingway, J. D., 1995. Practical Gamma-Ray Spectrometry. John Wiley & Sons
[63] Vogt, K., Persson H., 1991. Measuring growth and development of roots. In: Hincley, T., Lassoie, J.P. (Eds.), Techniques and Approaches in Forest Tree Ecophysiology, 477–501.
[64] Realo, K.; Koch, R.; Lust, M.; Uljas, A.; Realo, E., 2004. Lead-210 in air and surface soil in NE Estonia. In: Full Papers of IRPA-11, 11th Intern. Congress of Intern. Radiation Protection Association: 11th International Congress of International Radiation Protection Association; Madrid, Spain; 23–28 May, 2004. Spain:, 2004, 1–8.
[65] Preiss, N., Méliéres, M.-A., Pourchet, M., 1996. A compilation of data on lead 210 concentration in surface air and fluxes at the air-surface and water-sediment interfaces. J. Geophys. Res. 101: 28847–28862.
[66] Mustonen, R., 1985. Methods for Evaluation of Radiation from Building Materials. Radiation Protection Dosimetry 7, 235–238.
[67] Markkanen, M., 1995. Radiation Dose Assessments for Materials with Elevated Natural Radioactivity. Report STUK-B-STO32. Radiation and Nuclear Safety Authority – STUK.
[68] Isaksson, M., Erlandsson, B., 1998. Models for the vertical migration of 137Cs in the ground – A field study. J. Environ. Radioactivity 41, 163–182.
[69] Konchin, O.V., 1992. Mathematical model of 137Cs migration in soil: analysis of observations following the Chernobyl accident. Health Phys 63(3), 301–306.
[70] European Commission, 1995. Methodology for assessing the radiological consequences of routine releases of radionuclides to the environment. Report EUR 15760, Radiation Protection series no 72, Office for Official Publications of EC, Luxembourg.

PUBLICATIONS

CURRICULUM VITAE

Merle Lust

1.	Date of birth:	19. May 1974
2.	Citizenship:	Estonian
3.	Aadress:	Tähtvere 15–3, Tartu, Estonia
4.	Phone:	+3725174220
5.	E-mail:	merle.lust@gmail.com
6.	Current employer:	Eesti Teadusagentuur (Estonian Research Council), consultant
7.	Education:	Estonian School of Diplomacy, 2005, International Relations University of Tartu, 1999, MSc, Environmental Physics University of Surrey, 1998, MSc, Environmental and Radiation Protection University of Tartu, 1996, BSc, Physics and Environmental Physics
8.	Language skills:	Estonian, Russian, English, Spanish, Swedish
9.	Employment:	Estonian Research Council, 2012– SA Archimedes, 2010–2012, Eesti Energia 2009–2010, Estonian Radiation Protection Centre 2002–2009, University of Tartu, Insitute of Physics 1999–2002 Estonian Radiation Protection Centre 1996–1999

ELULOOKIRJELDUS

Merle Lust

1. Sünniaeg ja koht: 19. mai 1974
2. Kodakondsus: eesti
3. Aadress: Tähtvere 15–3, Tartu, Eesti
4. Telefon: 5174220
5. E-post: merle.lust@gmail.com
6. Praegune töökoht, amet: Eesti Teadusagentuur, koordinaator
7. Haridus (lõpetatud õppeasutused, lõpetamise aastad, omandatud kraadid, kvalifikatsioonid):
   Eesti Diplomaatide Kool, 2005, rahvusvahelised suhted
   Tartu Ülikool, 1999, MSc keskkonnafüüsika
   Surrey Ülikool, 1998, MSc keskkonna- ja kiirguskaitse
   Tartu Ülikool, 1996, BSc, füüsika-keskkonnafüüsika
8. Keelteoskus: eesti, vene, inglise, hispaania, rootsi
9. Töökogemus (teenistuskäik):
   Eesti Teadusagentuur 2012– ,
   SA Archimedes, 2010–2012,
   Eesti Energia 2009–2010,
   Kiirguskeskus 2002–2009,
   Tartu Ülikooli Füüsika Instituut 1999–2002,
   Kiirguskeskus 1996–1999.

DISSERTATIONES PHYSICAE UNIVERSITATIS TARTUENSIS

1. **Andrus Ausmees.** XUV-induced electron emission and electron-phonon interaction in alkali halides. Tartu, 1991.
2. **Heiki Sõnajalg.** Shaping and recalling of light pulses by optical elements based on spectral hole burning. Tartu, 1991.
3. **Sergei Savihhin.** Ultrafast dynamics of F-centers and bound excitons from picosecond spectroscopy data. Tartu, 1991.
4. **Ergo Nõmmiste.** Leelishalogeniidide röntgenelektronemissioon kiiritamisel footonitega energiaga 70–140 eV. Tartu, 1991.
5. **Margus Rätsep.** Spectral gratings and their relaxation in some low-temperature impurity-doped glasses and chrystals. Tartu, 1991.
6. **Tõnu Pullerits.** Primary energy transfer in photosynthesis. Model calculations. Tartu, 1991.
7. **Olev Saks.** Attoampri diapasoonis voolude mõõtmise füüsikalised alused. Tartu, 1991.
8. **Andres Virro.** AlGaAsSb/GaSb heterostructure injection lasers. Tartu, 1991.
9. **Hans Korge.** Investigation of negative point discharge in pure nitrogen at atmospheric pressure. Tartu, 1992.
10. **Jüri Maksimov.** Nonlinear generation of laser VUV radiation for high-resolution spectroscopy. Tartu, 1992.
11. **Mark Aizengendler.** Photostimulated transformation of aggregate defects and spectral hole burning in a neutron-irradiated sapphire. Tartu, 1992.
12. **Hele Siimon.** Atomic layer molecular beam epitaxy of A^2B^6 compounds described on the basis of kinetic equations model. Tartu, 1992.
13. **Tõnu Reinot.** The kinetics of polariton luminescence, energy transfer and relaxation in anthracene. Tartu, 1992.
14. **Toomas Rõõm.** Paramagnetic H^{2-} and F^+ centers in CaO crystals: spectra, relaxation and recombination luminescence. Tallinn, 1993.
15. **Erko Jalviste.** Laser spectroscopy of some jet-cooled organic molecules. Tartu, 1993.
16. **Alvo Aabloo.** Studies of crystalline celluloses using potential energy calculations. Tartu, 1994.
17. **Peeter Paris.** Initiation of corona pulses. Tartu, 1994.
18. **Павел Рубин.** Локальные дефектные состояния в CuO_2 плоскостях высокотемпературных сверхпроводников. Тарту, 1994.
19. **Olavi Ollikainen.** Applications of persistent spectral hole burning in ultrafast optical neural networks, time-resolved spectroscopy and holographic interferometry. Tartu, 1996.
20. **Ülo Mets.** Methodological aspects of fluorescence correlation spectroscopy. Tartu, 1996.
21. **Mikhail Danilkin.** Interaction of intrinsic and impurity defects in CaS:Eu luminophors. Tartu, 1997.

22. **Ирина Кудрявцева.** Создание и стабилизация дефектов в кристаллах KBr, KCl, RbCl при облучении ВУФ-радиацией. Тарту, 1997.
23. **Andres Osvet.** Photochromic properties of radiation-induced defects in diamond. Tartu, 1998.
24. **Jüri Örd.** Classical and quantum aspects of geodesic multiplication. Tartu, 1998.
25. **Priit Sarv.** High resolution solid-state NMR studies of zeolites. Tartu, 1998.
26. **Сергей Долгов.** Электронные возбуждения и дефектообразование в некоторых оксидах металлов. Тарту, 1998.
27. **Kaupo Kukli.** Atomic layer deposition of artificially structured dielectric materials. Tartu, 1999.
28. **Ivo Heinmaa.** Nuclear resonance studies of local structure in $RBa_2Cu_3O_{6+x}$ compounds. Tartu, 1999.
29. **Aleksander Shelkan.** Hole states in CuO_2 planes of high temperature superconducting materials. Tartu, 1999.
30. **Dmitri Nevedrov.** Nonlinear effects in quantum lattices. Tartu, 1999.
31. **Rein Ruus.** Collapse of 3d (4f) orbitals in 2p (3d) excited configurations and its effect on the x-ray and electron spectra. Tartu, 1999.
32. **Valter Zazubovich.** Local relaxation in incommensurate and glassy solids studied by Spectral Hole Burning. Tartu, 1999.
33. **Indrek Reimand.** Picosecond dynamics of optical excitations in GaAs and other excitonic systems. Tartu, 2000.
34. **Vladimir Babin.** Spectroscopy of exciton states in some halide macro- and nanocrystals. Tartu, 2001.
35. **Toomas Plank.** Positive corona at combined DC and AC voltage. Tartu, 2001.
36. **Kristjan Leiger.** Pressure-induced effects in inhomogeneous spectra of doped solids. Tartu, 2002.
37. **Helle Kaasik.** Nonperturbative theory of multiphonon vibrational relaxation and nonradiative transitions. Tartu, 2002.
38. **Tõnu Laas.** Propagation of waves in curved spacetimes. Tartu, 2002.
39. **Rünno Lõhmus.** Application of novel hybrid methods in SPM studies of nanostructural materials. Tartu, 2002.
40. **Kaido Reivelt.** Optical implementation of propagation-invariant pulsed free-space wave fields. Tartu, 2003.
41. **Heiki Kasemägi.** The effect of nanoparticle additives on lithium-ion mobility in a polymer electrolyte. Tartu, 2003.
42. **Villu Repän.** Low current mode of negative corona. Tartu, 2004.
43. **Алексей Котлов.** Оксианионные диэлектрические кристаллы: зонная структура и электронные возбуждения. Tartu, 2004.
44. **Jaak Talts.** Continuous non-invasive blood pressure measurement: comparative and methodological studies of the differential servo-oscillometric method. Tartu, 2004.
45. **Margus Saal.** Studies of pre-big bang and braneworld cosmology. Tartu, 2004.

46. **Eduard Gerškevitš.** Dose to bone marrow and leukaemia risk in external beam radiotherapy of prostate cancer. Tartu, 2005.
47. **Sergey Shchemelyov.** Sum-frequency generation and multiphoton ionization in xenon under excitation by conical laser beams. Tartu, 2006.
48. **Valter Kiisk.** Optical investigation of metal-oxide thin films. Tartu, 2006.
49. **Jaan Aarik.** Atomic layer deposition of titanium, zirconium and hafnium dioxides: growth mechanisms and properties of thin films. Tartu, 2007.
50. **Astrid Rekker.** Colored-noise-controlled anomalous transport and phase transitions in complex systems. Tartu, 2007.
51. **Andres Punning.** Electromechanical characterization of ionic polymer-metal composite sensing actuators. Tartu, 2007.
52. **Indrek Jõgi.** Conduction mechanisms in thin atomic layer deposited films containing TiO_2. Tartu, 2007.
53. **Aleksei Krasnikov.** Luminescence and defects creation processes in lead tungstate crystals. Tartu, 2007.
54. **Küllike Rägo.** Superconducting properties of MgB_2 in a scenario with intra- and interband pairing channels. Tartu, 2008.
55. **Els Heinsalu.** Normal and anomalously slow diffusion under external fields. Tartu, 2008.
56. **Kuno Kooser.** Soft x-ray induced radiative and nonradiative core-hole decay processes in thin films and solids. Tartu, 2008.
57. **Vadim Boltrushko.** Theory of vibronic transitions with strong nonlinear vibronic interaction in solids. Tartu, 2008.
58. **Andi Hektor.** Neutrino Physics beyond the Standard Model. Tartu, 2008.
59. **Raavo Josepson.** Photoinduced field-assisted electron emission into gases. Tartu, 2008.
60. **Martti Pärs.** Study of spontaneous and photoinduced processes in molecular solids using high-resolution optical spectroscopy. Tartu, 2008.
61. **Kristjan Kannike.** Implications of neutrino masses. Tartu, 2008.
62. **Vigen Issahhanjan.** Hole and interstitial centres in radiation-resistant MgO single crystals. Tartu, 2008.
63. **Veera Krasnenko.** Computational modeling of fluorescent proteins. Tartu, 2008.
64. **Mait Müntel.** Detection of doubly charged higgs boson in the CMS detector. Tartu, 2008.
65. **Kalle Kepler.** Optimisation of patient doses and image quality in diagnostic radiology. Tartu, 2009.
66. **Jüri Raud.** Study of negative glow and positive column regions of capillary HF discharge. Tartu, 2009.
67. **Sven Lange.** Spectroscopic and phase-stabilisation properties of pure and rare-earth ions activated ZrO_2 and HfO_2. Tartu, 2010.
68. **Aarne Kasikov.** Optical characterization of inhomogeneous thin films. Tartu, 2010.

69. **Heli Valtna-Lukner.** Superluminally propagating localized optical pulses. Tartu, 2010.
70. **Artjom Vargunin.** Stochastic and deterministic features of ordering in the systems with a phase transition. Tartu, 2010.
71. **Hannes Liivat.** Probing new physics in e+e− annihilations into heavy particles via spin orientation effects. Tartu, 2010.
72. **Tanel Mullari.** On the second order relativistic deviation equation and its applications. Tartu, 2010.
73. **Aleksandr Lissovski.** Pulsed high-pressure discharge in argon: spectroscopic diagnostics, modeling and development. Tartu, 2010.
74. **Aile Tamm.** Atomic layer deposition of high-permittivity insulators from cyclopentadienyl-based precursors. Tartu, 2010.
75. **Janek Uin.** Electrical separation for generating standard aerosols in a wide particle size range. Tartu, 2011.
76. **Svetlana Ganina.** Hajusandmetega ülesanded kui üks võimalus füüsikaõppe efektiivsuse tõstmiseks. Tartu, 2011
77. **Joel Kuusk.** Measurement of top-of-canopy spectral reflectance of forests for developing vegetation radiative transfer models. Tartu, 2011.
78. **Raul Rammula**. Atomic layer deposition of HfO_2 – nucleation, growth and structure development of thin films. Tartu, 2011.
79. **Сергей Наконечный.** Исследование электронно-дырочных и интерстициал-вакансионных процессов в монокристаллах MgO и LiF методами термоактивационной спектроскопии. Тарту, 2011.
80. **Niina Voropajeva.** Elementary excitations near the boundary of a strongly correlated crystal. Tartu, 2011.
81. **Martin Timusk.** Development and characterization of hybrid electro-optical materials. Tartu, 2012, 106 p.