

Environmental Radioactivity Measuring Methods

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Abstract Several aspects of environmental radioactivity measurements are surveyed. Starting with the development and goals of the surveillance of environmental radioactivity, the radionuclide coverage needed, the required sensitivities, and the materials to be analyzed are discussed. Presently used analytical and measurement methods are shortly reviewed. Emphasis is laid upon new and advanced techniques for both decay measurements and mass spectrometric methods. Because of the importance of environmental radioactivity measurements for regulatory issues, political decision making and for public discussions there is an extreme need for high quality data. Therefore, current issues of quality assurance and control, of international standardization and harmonization are dealt with in detail. Finally, the needs for dissemination of environmental radioactivity measurements in the public are discussed and possible future developments of environmental radioactivity measurements are outlined.

1 History and Goals of Environmental Radioactivity Measurements

The history of environmental measurements started with the onset of the nuclear age, in particular with the atmospheric nuclear weapons tests. It was driven by the concern of global radioactive pollution and of the resulting human radiation exposure. Compared to other sources, the by far largest amount of man-made radioactivity was emitted into the environment by the atmospheric nuclear weapons tests. This led to a contamination of atmosphere, hydrosphere, pedosphere and biosphere. The resulting radiation exposure of man reached 8 % of the natural exposure in beginning sixties (UNSCEAR, 1982)¹. Only after the Nuclear Test Ban Treaty came into effect in 1963 the undesirable situation started to improve. The legacy of the atmospheric explosions declined and presently the resulting individual radiation exposure is less than 10 $\mu\text{Sv/a}$ on the average (UNSCEAR, 2000).

A wealth of information was obtained in the fifties and sixties on environmental radionuclide abundances, radioecological processes and pathways of radionuclides through the environment to man. Systems of environmental radioactivity surveillance were installed in many countries and surveillance measurements, in particular of air and precipitation, became routine. National and international committees evaluated the findings and comprehensive reports exist. Exemplary, as a source of references of the past achievements, the series of UNSCEAR reports can be cited; see UNSCEAR (1988) for the historical development and UNSCEAR (2000) as the most recent one.

With the beginning of the peaceful use of nuclear energy and the installation of nuclear power plants environmental radioactivity measurements got a new quality. Because of the enormous radioactive inventories of power reactors the emissions from such installations and the resulting immissions into the environment had to be surveyed even during routine operation. Since such measurements had to be made against the natural radiation background and against the

¹ References in this paper are exemplary rather than comprehensive. They shall invite for further reading.

omnipresent fission and activation products of the atmospheric fall-out of the test explosions, the actual status of environmental contamination had to be measured prior to the operation of a nuclear installation followed by a permanent surveillance of both the emissions and the immissions in the surroundings of the plants. Incidents and accidents in the early phase of nuclear power use demonstrated the necessity of nation-wide surveillance of environmental radioactivity.

In Europe, the EURATOM Treaty (EURATOM, 1957) made this an international issue and basic guidelines for the protection of the public and of worker against the dangers of ionizing radiation were set (European Commission, 1959). The national authorities accepted the general responsibility to conform with these EURATOM basic radiation protection standards (European Commission, 1959, 1996a) and to perform a permanent surveillance of the radioactivity of air, water and soil and to inform the Commission about the results of this surveillance in compliance with articles 35 and 36 of the Treaty (European Commission, 2000b). A multitude of national systems of environmental surveillance was installed and a basis for a world-wide network of such installation was set into effect.

The public attitudes towards environmental radioactivity measurements were drastically changed by the Chernobyl accident in 1986. Differing attitudes in dealing with the Chernobyl fall-out in the European countries and lack of information from the former USSR demonstrated the necessity of a global dissemination of information on environmental radioactivity and of an international harmonization of its evaluation and assessment. Extending the existing surveillance systems, measures were taken in many countries to handle inhomogenous contamination also remote from nuclear installations, *e.g.* (BMU, 1986).

In the former USSR, large scale environmental radioactivity measurements were necessary in the regions highly contaminated by the Chernobyl accident. Decisions about evacuation of inhabitants and evaluations of the past, present and future radiation exposures had to be made on the basis of these environmental radioactivity measurements which later were internationally evaluated (IAEA, 1991). The consequences of the Chernobyl accident are still followed up and many independent assessments exist, *e.g.* (IAEA, 1996; NEA, 1995; SSK, 1996), and most recently by UNSCEAR (2000).

With the end of the cold war, further new tasks of environmental radioactivity measurements became evident which before were frequently kept secret on national levels. The legacy of the Cold War brought clear evidence of large scale contaminations and radioactive remainders; see UNSCEAR (2000) and IAEA (2000) for surveys and references. There were hitherto unknown major nuclear accidents such as the Cheliabinsk events. There was inadequate dumping of nuclear waste on land and in the sea and large contaminated areas due to Uranium mining and milling as *e.g.* in Ukraine, Kirgisistan, Kasachstan, France, Germany, and USA.

In particular from the military use of nuclear power, a large number of contaminated sites remained and were acknowledged by the public. These remainders also included in the Western world the vicinities of reprocessing and fabrication plants, *e.g.* (Wolbarst *et al.*, 1999), and the large-scale contamination of both the test grounds of atmospheric and underground nuclear explosions, *e.g.* in Kazakhstan (IAEA, 1998a), Bikini (IAEA, 1998b), Muroroa (IAEA, 1998c). In the military context, measurements of environmental radioactivity also became of imminent importance to detect nuclear weapons proliferation using the emissions of test explosions as well as of enrichment and reprocessing plants. These latter measurements need the outmost analytical sensitivity and isotopic accuracy since far-reaching conclusions have to be drawn from faintest traces in a bulk of environmental radioactivity.

Further tasks for environmental radioactivity measurements in the military field comprised measurements to describe the impact of the loss of nuclear warheads in plane crashes and of radionuclide energy systems of satellites which were set free by their destruction during reentry into the Earth's atmosphere.

Recently, also the question of battlefield surveillance became an additional public issue raising the question whether reliable experimental data can be obtained to describe or exclude enhanced exposure of the public and of soldiers after the use of ammunition containing depleted uranium (Rostker, 2000; Harley *et al.*, 1999, UNEP, 2000).

During the more than 4 decades of environmental radioactivity measurements, also emissions from technologies not related to nuclear power became issues. They included the routine release of ^3H and ^{226}Ra from industry manufacturing fluorescent dials or the inadequate disposal of such dials after World War II. Emissions of radionuclides from nuclear medicine hospitals today are found in municipal waste and waste waters and disposal of exempted radioactive waste in municipal waste storages or waste-burning facilities has to be surveyed. Accidental releases range from undesired melting of highly active nuclear sources in the steel industry, as *e.g.* in Taiwan (Woushou, 1997), to the dismantling of orphan sources as in the Juarez (Smith, 1984) and Goiana events (IAEA, 1988). Today an increasing number of orphan sources and of illicit trafficking is observed and environmental surveillance has to comprise detection and recovery of the dangerous material (UNSCEAR, 1982, 1988; Lubenau and Yusko, 1998).

The surveillance of natural radionuclides in the environment did not have high priority over many years compared to that of man-made radioactivity. There is, however, an increasing interest in such measurements since enhanced exposure to natural radioactivity is getting the same legal weight as any other radiation exposure. The recent EURATOM basic safety standards (European Commission, 1996a) explicitly state that also enhanced exposure to natural radioactivity and radiation is also an issue of radiation protection. In this context the surveillance of technologically enhanced naturally occurring materials, called TENORM or NORM, becomes important.

The historic development of the scopes of environmental radioactivity measurements can be summarized as a path through four different categories of measurements. It started from the surveillance of man-made radionuclides in the general territory of a country, went to that of man-made radionuclides at particular sites in the vicinity of (nuclear and other technical) installations, proceeded to the surveillance of natural radionuclides at particular sites and today ends with measurements of natural radioactivity in the general territories of countries.

Measurements of environmental radioactivity in all these four categories have multiple goals

- to provide a reliable experimental database to estimate the human radiation exposure,
- to proof the compliance of practices with radiation protection standards and legal exposure limits as well with international non-proliferation agreements,
- to record changes of environmental radioactivity and to establish a scientific basis on which human impact can be estimated with the goal to attain a sustainable development,
- to understand the behavior of radionuclides in the environment and their pathways to man,
- to exploit the potential of radionuclides in the environment as natural and man-made tracers of environmental processes and, last but not least,
- to give, beyond any doubt, evidence to the public about all facts of environmental radioactivity.

This wide variety of different goals needs various types of measurements, an extreme width of analytical techniques and methods with individual sensitivity requirements which will be discussed below.

2 Radionuclide Coverage, Sensitivity Requirements and Materials

A wide variety of radionuclides has to be analyzed in environmental materials. They cover fission products, transuranium nuclides, activation products and the naturally occurring radionuclides.

Though a few hundred different radionuclides are produced by the fission process, just a limited number of radionuclides with sufficiently long half-lives is of radiological significance and has to be covered by measurements of fall-out from atmospheric explosions and in surveillance of test sites and nuclear installations. These are the short-lived radionuclides ^{89}Sr , ^{95}Zr , ^{99}Mo , ^{103}Ru , ^{131}I , and ^{133}Xe , the medium-lived ones such as ^{90}Sr , ^{85}Kr , ^{106}Ru , $^{110\text{m}}\text{Ag}$, ^{137}Cs , ^{134}Cs , ^{124}Sb , ^{125}Sb , ^{154}Eu , and the long-lived nuclides ^{99}Tc , ^{129}I , ^{135}Cs . Of the transuranium radionuclides which are produced in nuclear chain reactors as well as in nuclear explosions ^{236}U , $^{238,239,240}\text{Pu}$, ^{241}Am , ^{243}Cm have to be covered. Activation of structural and shielding materials of fission reactors produces ^{54}Mn , ^{57}Co , ^{58}Co , ^{60}Co , $^{110\text{m}}\text{Ag}$ and activation of water and ambient air ^3H and ^{14}C . In future fusion reactors, ^3H and a variety of activation products will be the relevant radionuclides. With respect to the medium- and long-term storage of radioactive waste and also with respect to the disposal of exempted radioactive waste the surveillance of medium- and long-lived radionuclides will be of importance.

Considering radioactive waste, further activation products from accelerators, in particular from spallation neutron sources and possible future accelerator-driven facilities for transmutation of radioactive waste (Bowman *et al.*, 1992) and for energy amplification (Carminati *et al.*, 1993) have to be considered. According to our present knowledge, this adds the long-lived radionuclides ^{10}Be , ^{26}Al , ^{36}Cl , ^{41}Ca , ^{44}Ti , ^{53}Mn , ^{55}Fe , ^{60}Fe to the list of relevant radionuclides with respect to long-term disposal of radioactive waste.

According to the actual human radiation exposure of large populations, today the naturally occurring radionuclides, the natural and man-made variability of the exposure caused by these nuclides becomes of increasing interest. ^{222}Rn and ^{220}Rn and their decay products make up the bulk of natural radiation exposure and comprehensive measuring programs have been performed and are going on all over the world, see *e.g.* UNSCEAR (2000) for references. But, because of the limited space of this article we will not deal with this issue.

With respect to external and ingestive internal exposure to natural radionuclides from the geosphere, only the nuclides of the ^{232}Th , ^{235}U , and ^{238}U decay chains are relevant. They are of importance because of geologic anomalies, such as the famous monazite areas in India and Brazil or the ^{226}Ra anomalies in Iran, which may lead to exposure up to tens of mSv per year and might reach extremes up to 100 mSv/a. Further, they are the radionuclides which occur in technically enhanced naturally occurring radioactive materials which are problematic for the exposure of workers as well as of members of the public. Such materials occur in coal, gas and oil industries, in mining and milling of a variety of metals as well as in the exhaust gas of power plants burning fossil fuels. The use of naturally or technically enhanced radioactive materials as building and construction materials needs radioactivity measurements of the relevant radionuclides. The EURATOM basic safety standards (European Commission, 1996a) made surveillance of these isotopes and of the exposure to such materials a legal requirement.

While the external exposure can be easily determined since only the γ -emitting radionuclides are relevant here, for the description of the internal exposure one has to consider quite a number of radionuclides which have to be individually analyzed since radioactive equilibrium in nutrients cannot be anticipated. Since, moreover, the dose coefficients (ICRP, 1996) of the radionuclides of the natural decay chains show large differences, one needs for a reliable estimate of the internal exposure, measurements of the individual abundances in foodstuff and drinking water of ^{238}U , ^{234}U , ^{230}Th , ^{226}Ra , ^{210}Pb , ^{210}Po from the ^{238}U decay chain, ^{235}U , ^{231}Pa , ^{227}Ac , ^{227}Th , and ^{223}Ra from the ^{235}U decay chain and ^{232}Th , ^{228}Ra , ^{228}Th and ^{224}Ra from the ^{232}Th decay chain. For only a few of these nuclides radioactive equilibrium can be presumed in the hydro- and biosphere so that measurements of short-lived daughters is not needed to derive reliable data on the exposure. Such nuclides are e.g. ^{227}Th , ^{223}Ra and ^{224}Ra . As long as enriched or depleted Uranium can be excluded also the separate measurement of ^{235}U can be avoided. If this is, however, not the case, the utmost precision and isotopic accuracy is needed to determine all relevant uranium isotopes, namely ^{234}U , ^{235}U , ^{236}U and ^{238}U , as e.g. for non-proliferation control.

^{40}K adds only in rare cases to the external exposure of workers by NORM materials from KCl and NaCl mining. The internal exposure due to ^{40}K is homeostatically controlled depending on the physiological behavior of the essential trace element potassium rather than on its intake. From the other natural radionuclides in the geosphere only ^{87}Rb adds marginally to the human radiation exposure. But, there are not any radiological needs for measurements of this radionuclide. The same is true for all other natural primordial radionuclides.

From the cosmogenic radionuclides, only ^7Be and ^{14}C are of some, but minor importance with respect to radiation exposure and due to their relatively minor variability with variations of the galactic cosmic ray intensity over the solar 11-years cycle their measurement is only of interest in order to establish natural baseline data. The anthropogenic changes of ^{14}C will be discussed later.

Though technical or medical applications of radioactivity do not add further nuclides, these applications are also targets of environmental surveillance. Radionuclides such as ^{131}I from nuclear-medical therapy, ^{232}Th from welding electrodes, ^{60}Co , ^{137}Cs and ^{226}Ra from radionuclide sources, ^{235}U , ^{236}U , and ^{238}U from the technical and military use of depleted uranium which even may come from reprocessed nuclear fuel and may consequently contain also some impurities of transuranium elements (Rostker, 2000), and ^3H and ^{226}Ra from luminescent dials have to be taken into account.

Actual investigations or surveillance programs usually cover purpose-dependent subsets of all these radionuclides. Due to the multitude of purposes, however, for all these radionuclides measuring methods have to be available which have to satisfy adequate and reasonable sensitivity requirements.

When defining the sensitivity requirements of environmental radioactivity measurements one has to distinguish two extremes of purposes of such measurements, namely measurements which allow to determine exactly the present abundance of a nuclide in a particular environmental material, on the one hand, and measurements which have the goal to determine only those concentrations which have a defined radiological significance, on the other. These extremes can be categorized as scientific and surveillance measurements, respectively. Measurements to be performed for the purpose of non-proliferation control always will fall into the category of scientific measurements.

The radiological significance of an environmental radionuclide concentration usually is expressed in terms of an expected exposure due to this nuclide and the requirements are defined as required detection limits derived from radioecological models which connect the expected exposure with the concentration of the radionuclide in question. In the European reports on environmental radioactivity in the European Community, *e.g.* (European Commission, 1996b), the required detection limits are related to reporting levels for the surveillance of air, surface water, drinking water, milk and mixed total diet. These reporting levels RL are defined as

$$RL = \frac{DL}{RF \cdot EDC \cdot CF} \quad (1)$$

with

DL = annual dose limit, taken in accordance with ICRP (1991) to be 1 mSv,

RF = reduction factor of the dose limit, taken to be 1000,

EDC = effective dose coefficient according to ICRP (1996),

CF = annual consumption per person.

Using the thus defined reporting levels man-made radionuclides are surveyed at an environmental level which causes a risk of stochastic effects as low as $5 \times 10^{-8} \text{ a}^{-1}$ (European Commission, 1996b).

The sensitivity requirements of measurements of the actually existing radionuclide concentrations are, on the first glance, self-defined. However, they are reasonable only if naturally occurring radionuclides are to be measured because of their omnipresence. The actual concentrations of man-made radionuclides presently are so low in some environmental materials that it needs the most advanced measuring methods to detect them. Radionuclide concentrations in air can be taken as an example here where the activity concentrations of ^{60}Co , ^{90}Sr , ^{131}I and ^{137}Cs are nearly as low as 10^{-7} Bq/m^3 and about equal those of cosmogenic ^{22}Na . Naturally occurring radionuclides in air such as ^{40}K , ^{210}Pb and ^7Be are two, three and four orders of magnitude more abundant in air than ^{22}Na , respectively. Concentrations of ^{239}Pu and ^{240}Pu even are two orders of magnitude lower than those of ^{60}Co , ^{90}Sr , ^{131}I and ^{137}Cs . In view of such low concentrations measurements of man-made radionuclides at their actual levels appear not to be reasonable except for scientific purposes. Circumstances, however, which justify to perform such measurements also in the course of special surveillance measurements will become evident when dealing with the public opinion about radioactivity in the environment below. They are self-evident in the case of non-proliferation control.

The extreme range of radionuclide concentrations to be covered by environmental radioactivity measurements becomes even more evident if one considers that the total activity concentrations in air during the Chernobyl accident were between 1 and a few hundred Bq/m^3 in Western Europe and about equal to the activity concentrations in air of Radon decay products in Uranium-rich granite areas as *e.g.* in the Massif Central in France. Considering further that the Chernobyl accident did not cause a radiologically relevant enhancement of the radiation exposure in Western Europe and that measurements of environmental radioactivity also have to be applicable to accidental situations, where decisions about sheltering, evacuation and performance of thyroid blocking by administration of stable iodine have to be made, the orders of magnitude to be covered is further increased by a few decades.

The cited European report (European Commission, 1996b) and the organization of environmental radioactivity measurements according to the obligations of articles 35 and 36 of the EURATOM Treaty exactly distinguish the two extreme views of environmental radioactivity measurements by the organization of to different measuring networks, namely a dense and a

sparse network. The dense network spans over entire Europe to give sufficient information to detect and to quantify any significant radiation exposures due to man-made radionuclides on a local scale. The sparse network divides the present European Community in only 32 regions for each of which just one location is estimated to be sufficient to perform in-depth measurements of the actual concentrations of some selected radionuclides in order to describe the long-term trends of environmental radioactivity.

Reporting levels according to equation 1 are used and defined for the surveillance of environmental radioactivity in the European Community only for artificial radionuclides. The requirements of the EU basic standards (European Commission, 1996a) with respect to exposure to natural radioactivity will cause the necessity to define analogous required detection limits also for natural radionuclides. Examples for this do already exist as for instance in Germany where required detection limits are defined for the surveillance of the WISMUT remainders and for the remediation measures as a consequence of Uranium mining and milling (BMU, 1997, 1999).

There are, however, some general problems with the definition of required detection limits to be internationally applicable. First of all, the term detection limit has to be identically defined in the different countries. There is, however, no definition of the detection limit given in the cited EU Report (European Commission, 1996b) and looking *e.g.* for the EU Drinking Water Guideline (European Commission, 1998, 2000a) even contradictory definitions are given. We will discuss this point below when dealing with quality of measurements. Second, the required detection limits depend on the radioecological model used to connect radionuclide concentrations with dose. Presently used models, however, contain different degrees of conservative assumptions and thus make it difficult, if not impossible, to compare the required detection limits defined by different authorities. Third, the required detection limits depend on assumed consumption habits which differ from one country to another and which may contain again differing conservative assumptions. To overcome these difficulties in the international context will be a painstaking, but necessary work for the future. Moreover, there remains a historical distinction in perceiving the exposure to man-made and natural radionuclides, namely to use different measures for them.

We shall give an example for the need of harmonization. In Germany, the basis of the required detection limits for surveillance of man-made radionuclides according to the Act on the Precautionary Protection of the Population against Radiation Exposure (BMU, 1986) and to article 35 of the EURATOM Treaty uses a guideline value of the annual effective dose of 1 μSv which is in accordance with an annual dose limit DL of 1 mSv and a reduction factor RF of 1000 according to equation 1. For the surveillance of natural radionuclides from the WISMUT remainders (BMU, 1997) and for the surveillance of nuclear installations according to the Radiation Protection Ordinance (BMU, 1995a, 1995b) guideline values of the annual effective dose of 10 μSv are applied which are equivalent to a reduction factor RF of only 100. Given the amount of work and the costs induced by required detection limits, the harmonization of rules is essential for the surveillance of both, man-made and natural radionuclides.

An additional complication arises from the fact that in decommissioning of nuclear installation and exemption of contaminated or activated materials the expected radiation exposures of members of the public should not exceed a few time 10 μSv per year (UNSCEAR, 2000). Surveillance of such activities and of their consequences by environmental radioactivity measurements will need even more stringent sensitivity requirements.

As a final point of this chapter, the materials to be analyzed and the frequency by which these materials have to be surveyed shall be shortly discussed. As wide the variety of purposes of environmental radioactivity is, as differing are the requirements for materials to be analyzed, sample numbers and sampling frequencies. Given the costs induced in each actual investigation or surveillance program these requirements have to be scrutinized. If the ultimate purpose is the determination of the radiation exposure there are different ways to address this problem. The main distinction of approaches is whether they make use of reference persons or of really existing ones representing a critical group. While the approach using reference persons has been successfully applied in planning of practices and installations it appears to be hardly feasible to determine the actual radiation exposure of existing installations realistically by it. Even, if the results of environmental radioactivity measurements of a wide variety of food-stuffs is used as input to respective model calculations using *e.g.* models as ECOSYS (Müller and Pröhl, 1993) only exposure averages of normally behaving persons can be determined with some confidence. The problem rests then with non-normal behavior of certain individuals and with their lifestyle and consumption habits. Even if one excludes extremes or pathological behavior large uncertainties remain for the determination of the individual exposure. If averages of the exposure are sufficient the use of reference persons is adequate. The cited EU report (European Commission, 1996b) makes use of this reference-person concept with a minimum coverage of materials to be analyzed, namely air, drinking water and mixed total diet.

Another approach in the context of the exposure of a reference person is the use of aggregated transfer factors in a fall-out situation based on the analysis of soil and the determination of deposition densities. Thus, by a minimum of materials analyzed, the exposure can be estimated. In this case, the radioecological model used to determine the aggregated transfer factors is decisive and again the extend to which conservative assumptions are included.

Individual and realistic exposures can only be determined using the critical group concept evaluating the actual exposure of actually existing people. To determine the effective annual dose $E(t_1)$ for a year t_1 , this requires to quantify its contributions due to external exposure E_{ext} and due to ingestion and inhalation of radionuclides, E_{ing} and E_{inh} . According to ICRP 60 (ICRP, 1991) or the EURATOM Basic Safety Standard (European Commission, 1996a) such an evaluation has to be performed for the different tissues T taking into account the age dependence of exposure and of dose factors and the tissue weighing factors w_T .

$$E = E_{\text{ext}} + E_{\text{ing}} + E_{\text{inh}} = \sum_T w_T \cdot (E_{T,\text{ext}} + E_{T,\text{ing}} + E_{T,\text{inh}}) \quad (2)$$

Frequently, it is convenient to distinguish for the ingestion exposure path consumption of drinking water and of nutrients so that the annual effective dose for a tissue is given by

$$E_T(t_1) = E_{T,\text{ext}}(t_1) + E_{T,\text{inh}}(t_1) + E_{T,\text{ing,w}}(t_1) + \sum_n E_{T,\text{ing,n}}(t_1) \quad (3)$$

with

$E_T(t_1)$: annual effective dose in tissue or organ T during the year t_1 in Sv

$E_{T,\text{ext}}(t_1)$: annual effective dose in tissue or organ T due to external exposure to airborne radionuclides and to radionuclides in or deposited on the ground during the year t_1 in Sv

$E_{T,\text{inh}}(t_1)$: annual effective dose in tissue or organ T due to inhalation during the year t_1 in Sv

$E_{T,\text{ing,w}}(t_1)$: annual effective dose in tissue or organ T due to consumption of drinking water during the year t_1 in Sv

$E_{T,\text{ing,n}}(t_1)$: annual effective dose in tissue or organ T due to consumption of a nutrient n during the year t_1 in Sv

The external exposure due to γ -radiation is given by

$$E_{T,a}(t_1) = c \cdot \int_{t_1} \dot{H}_X(t) dt \quad (4)$$

with

$\dot{H}_X(t)$: ambient γ dose rate as a function of time t for the actual locations

c : conversion factor in Sv/Gy.

Analogously, the exposure of the skin due to β -immersion of airborne radioactivity can be evaluated. Due to the limited space we will not deal further with the external exposure.

The internal exposure due to inhalation is given by

$$E_{T,\text{inh}}(t_1) = \sum_r E_{T,\text{inh},r}(t_1) = \sum_r \sum_{t=0}^{t_1} A_r(t) \cdot \dot{V}(t) \cdot \int_{t_1-t}^{t_1-t+1} \frac{\partial g_{\text{inh},r,T}}{\partial t} dt \quad (5)$$

with

r : radionuclide in question

$A_r(t)$: activity concentration of the radionuclide r in air during a year t in $\text{Bq} \cdot \text{m}^{-3}$

$\dot{V}(t)$: age dependent breathing rate during a year t in $\text{m}^3 \cdot \text{s}^{-1}$

$g_{\text{inh},r,T}$: age dependent dose factor for tissue or organ T due to inhalation of a radionuclide r in $\text{Sv} \cdot \text{Bq}^{-1}$ according to ICRP (1996).

Note that for the intake of radionuclides with long physical and biological half-lives the exposure in a year t_1 contains contributions from previous years and may not be in equilibrium. In particular with respect to the exposure to some long-lived radionuclides of the natural decay series this is of importance. It is accounted for in equation 5 by summing over the entire lifetime up to year t_1 and by using the time differentials of the dose factors. This holds also for the exposure due to ingestion given below.

The internal exposure due to consumption of drinking water is given by

$$E_{T,\text{ing,w}}(t_1) = \sum_r E_{T,\text{ing},r,w}(t_1) = \sum_r \sum_{t=0}^{t_1} C_r^w(t) \cdot U^w(t) \cdot \int_{t_1-t}^{t_1-t+1} \frac{\partial g_{\text{ing},r,T}}{\partial t} dt \quad (6)$$

with

- $C_r^w(t)$: time dependent activity concentration in drinking water of a radionuclide r during the year t in $\text{Bq} \cdot \text{l}^{-1}$
- $U^w(t)$: age dependent consumption of drinking water during a year t in l
- $g_{\text{ing},r,T}$: age dependent dose factor for tissue or organ T due to ingestion of a radionuclide r in $\text{Sv} \cdot \text{Bq}^{-1}$ according to ICRP (1996).

The internal exposure due to consumption of nutrients is given by

$$E_{T,\text{ing},n}(t_1) = \sum_r E_{T,\text{ing},r,n}(t_1) = \sum_r \sum_n \sum_{t=0}^{t_1} C_r^n(t) \cdot U^n(t) \cdot \int_{t_1-t}^{t_1-t+1} \frac{\partial g_{\text{ing},r,T}}{\partial t} dt \quad (7)$$

with

- $C_r^n(t)$: time dependent activity concentration in a nutrient n of a radionuclide r during a year t in $\text{Bq} \cdot \text{kg}^{-1}$
- $U^n(t)$: age dependent consumption of a nutrient n during a year t in kg

For an as-realistic-as-possible determination of the dose, integrals over the time differentials of the dose factors are used in equations 5 - 7 in order to account properly for the built-up of the exposure due to long-lived radionuclides with long biological half-lives as they occur e.g. in the natural decay series. In the latter cases a conventional calculation of internal exposure, e.g. for ingestion by equation 8 on the basis of 50- or 70-years dose factors $g_{\text{ing},r,T}$ according to ICRP (1996), would strongly overestimate the dose committed between years t_0 and t_1 , in particular for children in their first years.

$$E_{T,\text{ing},n}(t_1) = \sum_r E_{T,\text{ing},r,n}(t_1) = \sum_r \sum_n C_r^n(t_1) \cdot U^n(t_1) \cdot g_{\text{ing},r,T} \quad (8)$$

For all radionuclides with short physical or biological half-lives, however, equation 8 also holds for a realistic assessment of the committed dose and for them also equations 5 and 6 can be simplified accordingly.

The determination according to equations 2 - 7 of an actual exposure of an individual member of the public is extremely costly and time-consuming. The situation becomes even worse if the distribution of effective doses in a population as well as the expectation value of such distributions shall be determined by the approach described here. Therefore, it is mainly used in the context of the critical group concept. Here, the recent investigation of exposure from emissions from La Hague (Nord-Cotentin Radioecology Group, 1999, 2000) can be taken as an outstanding example.

For an assessment of the human exposure environmental radioactivity measurements have to provide a complete and reliable data-set of the concentrations of natural and man-made radionuclides in air, water, soil and nutrients. If extremely low exposures are excluded on the basis of a *de minimis* concept certain radionuclide concentrations can be excluded as not relevant.

There are no *de minimis* if environmental radioactivity measurements are related to proliferation control. Such measurements have to distinguish faintest traces of elemental and/or isotopic patterns hidden under the bulk of environmental radioactivity. For them, methodologies for ultra-trace analysis at the pg- and fg-levels have to be applied which comprise the use of clean rooms and highest-purity reagents for sample preparations and chemistry. Even then, they can only be successful if the outmost blank and contamination control is performed. It is, however, a particular advantage for such measurements that unambiguous isotopic signatures of uranium and transuranium elements allow to distinguish the relevant components within the ubiquitous environmental radioactivity. Comparable analogues from other fields of sciences or applications can be found in geo- and cosmochemistry where analyses by isotopic dilution are common and mass spectrometric analysis of long-lived radionuclides and of actinides in radioactive disequilibrium are used for dating purposes. Just in rare cases, e.g. if claims of particular exposures to transuranium radionuclides emitted from nuclear installations have to be investigated against the background of nuclear weapons fall-out, such analyses are also necessary in the context of the determination of the human radiation exposure.

3 Methods: From counting decays to counting atoms

During five decades of environmental radioactivity measurements an outstanding evolution of measuring methods took place. Decay counting evolved from single channel counting over coincidence techniques with just a few counting channels to more or less sophisticated α -, β -, and γ -spectrometry with systems coupled to multi-channel analyzers. In parallel, the evolution of mass spectrometric methods made it possible to measure the low isotopic abundances of radionuclides in the presence of stable isotopes or isobars many orders of magnitude more abundant than the radionuclides themselves. Also general trends of technology development contributed to the improvement of methods as the increasing computerization and miniaturization which made it feasible to handle huge amounts of data and to use the most sophisticated methods for their evaluation. The evolution of analytical and radiochemistry strongly influenced the methodology of environmental radioactivity measurement. Use of clean rooms, high-purity reagents, strict contamination and blank control and impressive advances of chromatographic methods have to be mentioned here. Here, the old and today partially obsolete methods will just shortly be mentioned. Rather, the present state of the art will be discussed in this chapter emphasizing some outstanding developments.

Lacking more refined methods, the onset of decay counting made use of simple measurands such as gross alpha and gross beta activity concentrations in the various environmental materials to be analyzed. Large volume counters were developed with strong low-activity passive shielding and with active suppression of the cosmic ray background by coincidence and anti-coincidence shields. These low-level counters developed into various directions by the improvement of electronics in particular of coincidence techniques. Measurands such as residual beta activity concentration allowed to coarsely discriminate radiation from artificial radionuclides against that natural radionuclides from the decay series. Measurements of gross beta activity (as ^{90}Sr -equivalent) or gross alpha activity (as Pu-equivalent) are still frequently performed though no longer state-of-the-art.

Today, many of these techniques and their respective measurands are obsolete. The only justification of their application is in measurements which simply are intended to exclude the presence of any relevant α - or β -activity. Even if they are used for such purposes as prescribed for instance in the annex of the new drinking water guideline (European Commission, 1998, 2000a) more advanced nuclide-specific methods have to be used if certain limits which are considered as normal or harmless are exceeded. Having in mind the main goal to quantify the human exposure by measurements of environmental radioactivity and considering the

enormous differences in dose coefficients for different α - or β -decaying radionuclides, measurements such as gross alpha or gross beta or residual beta activity concentrations do not satisfy any need of an adequate dose assessment.

Today, nuclide specific data are needed in any case and they can be obtained from decay measurements either by single or few channel counting measurements after chemical separation of a particular radionuclide or by spectrometric measurements. Thus, counting measurements still are relevant for the analysis of certain radionuclides such as ^3H , ^{14}C , and ^{90}Sr though also in these cases other methods are strongly competing and even superior. For the measurement of β -decaying radionuclides the development of liquid scintillation spectrometric methods was the break-through to nuclide-specific measurements. Liquid scintillation methods evolved from single- or few-channel counting systems into multi-channel spectrometric ones and find wide-spread application. This applies also to the analysis α -emitting radionuclides. Alpha liquid scintillation spectrometry with β/γ discrimination has become a quick, reliable and low cost method with high sensitivity (~ 0.1 mBq). For a survey on recent developments of liquid scintillation spectrometry see (Cook *et al.*, 1996).

For α - and γ -spectrometry, it was the development of solid state semi-conductor detectors made of silicon or germanium which opened the field of nuclide-specific measurements. α -spectrometry with Si detectors still needs highly sophisticated chemical separation schemes to extract the radionuclides from the matrix and to prepare thin samples to avoid self-absorption and strong peak tailing due to stopping of the α -particles in the sample. In contrast, γ -spectrometry made possible non-destructive measurements of a wide range of relevant radionuclides.

Just for completeness it is to mention that for γ -radiation the evolution from counting to spectrometry passed the phase of solid state scintillation detectors having high counting efficiency but poor energy resolution. Though the resolution of plastic or NaI(Tl) scintillation detectors is much worse than that of semiconductor detectors they have maintained the importance for certain applications such as measurements of ambient dose rates, distinction of natural and artificial radionuclides in field measurements and in spectrometric systems where they serve in combination with semi-conductor detectors as active shields or for the suppression of the Compton background. Also coincidence techniques which were successful as low-level counters using γ - γ -coincidence or triple- γ -coincidence techniques (Herpers *et al.*, 1969) developed into highly sophisticated γ - γ -coincidence spectrometry, *e.g.* (Perkins *et al.*, 1970), which can be regarded as one extreme of low-level γ -spectrometry and which today are used in quite a number of laboratories.

The development of γ -spectrometric methods was strongly influenced by three aspects. First, the availability of intrinsically pure germanium allowed improvements of energy resolution and the construction of large volume germanium detectors which reached the same efficiency as NaI(Tl) detectors and of dedicated detectors optimized for the measurement of low-energy X- or γ -radiation or of high-energy γ -radiation. The improvements of detector size and of specialized design is, however, accompanied by complications in the evaluation of γ -spectra which may need corrections for systematic sum coincidences and complicated methods of efficiency calibration. For a general discussion of related problems see for instance (Debertin and Helmer, 1988).

Second, the improvement of detector shielding and of the reduction of natural radioactivity of the detector itself and the surrounding components. The improvement of detector shielding

included the use of low-activity lead shields, inner lining of the lead shields by other metals to reduce cosmic ray neutrons and to shield the radiation from their capture γ -rays and from the γ -rays of ^{210}Pb inside the lead shield. Methods of reducing γ -background from Radon decay products inside the shield range from the reduction of the free volume inside the shield by *e.g.* paraffin, to float the interior by absolutely filtered air or by high-purity nitrogen. Considerable improvements were made over the years due to the development of material sciences in particular with respect to the production of high-purity material which allowed to select and use low-activity components only and to construct special low-level detectors.

Third, the application of active shields and of electronic components which make use of coincidence and anti-coincidence techniques allowed to reduce the Compton background and, to a certain degree, also of the cosmic ray background. The presently final and decisive step of low-level γ -spectrometry is, however, the combination of all these methods described above and to go with the laboratory into the deep underground. to reduce the hadronic component of the cosmic radiation and to avoid both the continuum of cosmic ray background and the $^{74}\text{Ge}(n,n'\gamma)$ and $^{72}\text{Ge}(n,n'\gamma)$ background. Compared to an unshielded detector at sea level, the background of a γ -spectrometer can be reduced by three orders of magnitude and more in a deep-underground laboratory by application of these combined methods. For a detailed discussion of these aspects see for instance (Heusser, 1995).

A contrary approach of measuring environmental γ -emitting radionuclides is to use no shielding at all and to directly assess the abundances of radionuclides by field measurements via *in situ* γ -spectrometry.

With these technological developments, the state of the art of radioactivity measurements can be described as follows. For all γ -emitting radionuclides γ -spectrometry with Ge-detector is the method of choice. For the analysis by γ -spectrometry of radionuclides from the natural decay chains, however, a careful selection of γ -energies used for evaluation and the consideration of and correction for interfering γ -lines is important, *e.g.* (Schkade *et al.*, 1999). Moreover, it is essential to attain radioactive equilibrium in the samples to be measured since many radionuclides are determined via the γ -rays of their decay products. This holds for ^{238}U , ^{226}Ra , ^{227}Ac , and ^{228}Th which are determined via the γ -rays of ^{234}Th or $^{234\text{m}}\text{Pa}$, by ^{214}Pb or ^{214}Bi , by ^{227}Th , and by ^{224}Ra , ^{212}Pb or ^{208}Tl , respectively. Further, measurements different detectors (high- and low-energy) are partially needed, *e.g.* to obtain reliable results for ^{210}Pb . For the low-energy γ -radiation of this nuclide the determination of the counting efficiency in large volume samples is difficult and may need an additional measurement with a small sample by a low-energy detector.

For radionuclides emitting exclusively α - or β -radiation non-destructive methods cannot be applied and chemical or physical-chemical separation from the matrix are needed combined with skillful procedures for the preparation of the radionuclide samples to be measured. We will discuss the required separation techniques only in some special cases. Else we just mention some references from a broad literature of evaluated separation schemes available and prescribed by national authorities for the respective analyses (AFNOR, 2000; AKU, 1999; BMU, 1998; DoE, 1979). These evaluated schemes mostly make use of well established chemical techniques and usually recognize new analytical developments only at late stages of maturity. New developments and techniques such as modern chromatographic methods, as *e.g.* capillary electrophoresis, ionic chromatography, and HPLC, are scarcely found in these schemes today. Just becoming state-of-the-art in scientific investigations of environmental radioactivity and in proliferation control, modern chromatographic technique provide a wealth

of possible improvements, in particular with respect to automation of separations and to suppression of interferences. Independent of the application of modern or old-fashioned chemical techniques, radiochemical separations remain the prerequisites for the determination of α - and β -decaying radionuclides by consequent counting or spectrometry.

For ^{14}C , the analyses generally comprise the separation as CO_2 followed by measurement in gas-filled counters, by liquid scintillation counting or spectrometry or by accelerator mass spectrometry which will be discussed below in some detail. For ^{85}Kr counting techniques using gas-filled counting tubes into which the Krypton extracted from the air is included is still the method of choice. For ^3H , ^{89}Sr , ^{90}Sr , and ^{99}Tc , liquid scintillation counting or spectrometry is the method of choice after separation or enrichment though counting measurements with gas-filled detectors still remain a practicable option.

A particular problem arises in case of the analysis of ^{90}Sr (and ^{89}Sr). Here, all counting techniques have the disadvantage that after separation the built-up of the ^{90}Sr decay product ^{90}Y ($T_{1/2} = 64.1$ h) has to be waited for. The delay of more than one week between chemical separation and counting is not acceptable in case of a nuclear accident. For instance, after the Chernobyl accident there was too long an uncertainty about the contribution of fall-out ^{90}Sr in Western Europe for the decision makers. Considerable efforts have since been undertaken in order to develop fast methods for the determination of ^{90}Sr in case of nuclear emergencies when it is also required to deal with the counting-interfering presence of ^{89}Sr ($T_{1/2} = 50.5$ d). To accelerate the chemical separation of ^{90}Sr advantage has been taken from the development of special extraction reagents such as crown-ethers which, however, do not decrease the waiting time between separation and counting (Tait and Wiechen, 1997). Except for the mass spectrometric methods discussed below, liquid scintillation β -spectrometry can be applied to solve this remaining problem. β -spectrometry allows to distinguish ^{90}Sr , ^{90}Y and ^{89}Sr so that within less than two days after sampling a decision measurement can be made as to what degree consumption decisions are needed (Filb *et al.*, 1998).

β -decaying radionuclides of the natural decay series generally do not pose a problem since they all can be analyzed by γ -spectrometry of their decay products though in some cases chemical separations have to precede the measurements.

Counting and spectrometric methods, even in combination with chemical separations, come to their limits if long-lived radionuclides with half-lives exceeding 1000 a have to be measured. In principle, it would be much more advantageous to count the atoms of those radionuclides instead of waiting for their decay. Thus, mass spectrometric methods are the obvious choice. However, conventional mass spectrometry was and still is not able to detect the spurious amounts of long-lived radionuclides in the presence of stable isotopes or isobars which might be more abundant by more than 10 orders of magnitude. In addition, conventional mass spectrometry has to struggle with molecular interferences which may mimic the presence of a radionuclide. Therefore, conventional mass spectrometry in form of classical ICP-MS was applied successfully to environmental radioactivity measurements only in case of uranium and thorium where the problem is not the isotopic analysis but the analytical blank.

Only recently, new developments in mass spectrometry have opened up a new world of environmental radioactivity measurements. Extreme sensitivity and suppression of isotopic and isobaric interferences were successfully realized by accelerator mass spectrometry (AMS), by multi-collector magnetic sector field ICP-MS and by laser resonance ionisation mass spectrometry (RIMS). In case of AMS and ICP-MS, environmental radioactivity measurements have widely profited from analytical needs of geo- and cosmochemistry and still today the

analytical capabilities used in these fields of sciences are not yet fully exploited for environmental radioactivity measurements.

In accelerator mass spectrometry (AMS), the problem of isobaric, isotopic and molecular interferences can be avoided for long-lived radionuclides such as ^{10}Be , ^{14}C , ^{26}Al , ^{36}Cl , ^{41}Ca , ^{53}Mn , ^{59}Ni , ^{60}Fe , ^{90}Sr , ^{99}Tc , ^{129}I , and ^{236}U . Also for AMS, the radionuclide in question has to be chemically separated with or without addition of a stable isotopic carrier and a sample for measurement has to be prepared.

The AMS measurement is then performed with a nuclear physics accelerator mostly a tandem accelerator with a few MV terminal voltage. From the sample, negative molecular or atomic ions are produced in a Cs sputter ion source. The extracted ions are analyzed by magnetic and electric deflection and injected into the accelerator where they are accelerated towards the HV terminal. At the terminal a thin gas target or a thin stripper foil transforms the negative ions into positive ones and, at the same time, destroys any molecules in the beam. The positive ions are accelerated again to ground potential. When leaving the accelerator a suitable charge state of the ions is selected and by beam-forming electric and magnetic analyzers isobaric interferences are suppressed. The high energies of the ions now allow to make use of detection methods of nuclear physics which distinguish Z and A of the detected particles. Applying time-of-flight methods and ΔE -E detectors isotopic interferences are suppressed and isotopic ratios of the radionuclide and a neighboring isotope of down to 10^{-15} can be measured. For a detailed review and recent achievements see (Finkel and Suter, 1993) and the proceedings of the last AMS conference (Kutschera *et al.*, 2000).

Presently, AMS measurements are performed at a considerable number of AMS facilities all around the world. The disadvantage of AMS using huge accelerator systems decreased during recent years and dedicated smaller machines have been constructed. Just recently, an extremely small AMS machine has been developed (Suter *et al.*, 2000) so that a future of AMS as a wide-spread laboratory method comes into view.

AMS was successfully applied to the measurement of long-lived cosmogenic nuclides in terrestrial and extraterrestrial matter; see (Michel, 1999) for a review. With respect to the analysis of man-made radionuclides in the environment, its capabilities are just at the beginning to be fully exploited. For the radionuclides ^{14}C , ^{36}Cl , and ^{129}I , AMS offers the possibility to investigate the long-term impact of the peaceful and military use of nuclear fission and fusion. In case of ^{14}C , the activities of modern carbon can also be analyzed by low-level counting methods and the question which method is advantageous is decided by the time needed for analysis and how low $^{14}\text{C}/\text{C}$ ratios are to be covered. In using ^{14}C as an environmental tracer, low-level counting quickly comes to its limits and AMS is the method of choice.

It should be emphasized that AMS is also the method of choice for all the long-lived radionuclides produced as activation products at accelerators, spallation neutron sources and possible future accelerator-driven devices. In this context, also recent and ongoing measurements of long-lived activation products in materials from Hiroshima and Nagasaki have to be mentioned which are measurable by AMS with the purpose to resolve still existing discrepancies in the evaluation of the contributions of neutrons and γ -radiation to the exposure resulting from the explosions of nuclear weapons at Hiroshima and Nagasaki (Rühm *et al.*, 1998).

In order to cover both natural and man-made abundances of ^{36}Cl and ^{129}I only AMS is capable to cover the entire range of isotopic ratios observed in nature. ^{36}Cl showed an impressive bomb peak up to three orders of magnitude above normal, which, by now, nearly has decayed

to normal ratios, the bomb produced ^{36}Cl being diluted by marine Cl and washed out of the atmosphere (Synal *et al.*, 1990, 1997). Also the activities of ^{14}C which in the beginning of the sixties went up to 200 percent of the activity of modern pre-nuclear carbon nearly has returned to normal values, the biospheric response becoming normal on a time scale of 15 a (Levin *et al.*, 1997; Nakamura *et al.*, 1987).

The situation is more complicated for the long-lived radionuclide ^{129}I ($T_{1/2} = 15.7$ Ma). Since the middle of our century the environmental levels of ^{129}I with $^{129}\text{I}/^{127}\text{I}$ ratios of $\sim 10^{-12}$ have been dramatically changed as a consequence of the civil and military use of nuclear fission; see Schmidt *et al.* (1998) for a review. From the radioecological point of view, our knowledge about ^{129}I in the environment is still marginal. In 1962, radiochemical neutron activation analysis (RNAA) became available as a first analytical method to determine ^{129}I in environmental samples (Studier *et al.*, 1962). Manifold analyses demonstrated the extreme changes as a consequence of atmospheric weapon tests and even more pronounced due to releases from reprocessing plants. But, as discussed by Schmidt *et al.* (1998) in detail, RNAA is only capable of measuring $^{129}\text{I}/^{127}\text{I}$ ratios above 10^{-10} and the natural abundances and their transition to modern high contamination levels could not be quantified. Only by accelerator mass spectrometry it became possible to determine all $^{129}\text{I}/^{127}\text{I}$ ratios occurring in nature.

Moreover, many of the early analyses suffered from methodological deficits such as missing blank analyses and quality control. Although the number of ^{129}I investigations increased dramatically in recent years, the radioecology of ^{129}I is still insufficiently known. We therefore started a systematic investigation to establish reliable analytical for ^{129}I and ^{127}I analysis in various environmental materials and to close some gaps in our knowledge about the environmental abundances of ^{129}I , about its radioecological behavior and to exploit the potential of ^{129}I for the retrospective dosimetry of ^{131}I exposure after a nuclear accident such as the Chernobyl accident protocols (Schmidt *et al.*, 1998; Szidat *et al.*, 1999, 2000a, 2000b).

The results demonstrate that the natural abundances of ^{129}I have been sustainably changed and that there is a continuing increase of anthropogenic ^{129}I in the pedosphere and the terrestrial biosphere in Europe. Though the environmental ^{129}I levels are not of radiological concern at present, the future development should be carefully monitored. Generally, ^{129}I , together with ^{14}C and ^{36}Cl , can be regarded as excellent tools to quantify the long-term impact of nuclear power onto the environment.

It has to be mentioned that in situations with high ^{129}I releases also simpler methods as AMS and RNAA can be sufficiently sensitive to determine environmental ^{129}I abundances. So, Frechou *et al.* (2000) demonstrated that direct γ -X spectrometric measurements are possible *e.g.* in the vicinity of the La Hague reprocessing plant. These authors also developed new biospheric reference materials for the determination of ^{129}I and validated it using as three independent methods direct γ -X spectrometric measurements, RNAA and AMS. Tait *et al.* (2000) showed that also fast chemical separations with subsequent γ -spectrometric measurements by low-energy Ge-detectors can be sufficiently sensitive to reach the required detection limits for the surveillance of radioactive waste depositories in case of accidental releases.

The second new-frontier mass spectrometric method for environmental radioactivity measurements is ICP-MS. It has been successfully used for many years for trace element analysis. Also for ICP-MS, radiochemical separations have to be applied to avoid elemental interferences and a careful control respectively determination of the chemical yield is necessary. The sensitivity is in the fg-range, but molecular, isobaric and isotopic interferences remained crucial. Recently, however, ICP-MS got a new quality which probably will have great impact on

environmental radioactivity measurements. With ICP coupled to magnetic-sector-field multiple-collector mass spectrometry, e.g. (Halliday *et al.*, 1995), the problems of classical ICP-MS were widely overcome and, at the same time, the sensitivity was significantly improved. In geo- and cosmochemistry, this allowed to exploit radioactive disequilibria for dating purposes with an unprecedented accuracy (Halliday *et al.*, 1998). Disequilibrium dating using the uranium and thorium decay series, e.g. (Stirling *et al.*, 2000), are closely related to environmental radioactivity measurements and the potential of the new technique for environmental radioactivity measurements is striking. It was successfully applied to distinguish the remainders of depleted uranium ammunition in the presence of omnipresent natural uranium (UNEP, 2000). Generally, it can be applied to high precision actinide isotopic analyses and measurements of transuranium elements, as e.g. plutonium in materials with extreme Pu/U ratios below 10^{-14} , are essential for migration studies and for proliferation control. Measurements by the new ICP-MS techniques also will strongly facilitate comprehensive surveillance of long-lived radionuclides from the natural decay series in water and foodstuffs with the purpose of a better understanding of the pathways of the heavy radioelements to man and of the resulting radiation exposure. The new ICP-MS technique has, moreover, a large potential for automation by direct coupling of the analytical detection system with the radiochemical separation using capillary electrophoresis, ionic chromatography, HPLC.

In view of the trend to use mass spectrometric methods in environmental radioactivity measurements resonance ionisation mass spectrometry (RIMS) has to be mentioned as a third outstanding development. For this method the carrier-free element is chemically separated and measured by a RIMS set-up in which a time-of-flight mass spectrometer is fed by ions from a laser ion source. The suppression of undesired particles in the mass spectrometer is achieved by ionizing only atoms of the desired element respectively isotope by multiple-resonant laser excitation and ionisation. The new method has an extremely high element sensitivity and is well suited for trace analysis of long-lived radionuclides in environmental, biological and technical samples. It has the same superiority compared to counting measurements as discussed above for AMS and ICP-MS.

RIMS has been successfully used to analyze ^{41}Ca , ^{89}Sr , ^{90}Sr , ^{99}Tc , and Pu-isotopes at environmental abundances and it is applicable also for the analysis of Cm, Bk and Cf (Wendt *et al.*, 1997a, 1997b, 1999; Erdmann *et al.*, 1997; Eichler *et al.*, 1997; Müller *et al.*, 2000; Passler *et al.*, 1997). A special advantage of RIMS of plutonium isotopes is that the isotopic composition of all isotopes can be determined with an unprecedented accuracy with detection limits down to 10^6 atoms per sample.

4 Quality: Not just a Matter of Taste

It cannot be taken for granted that every measurement result has high quality and is reliable. A permanent struggle for precision and accuracy is characteristic for any experimental research. But, erroneous, low-quality, false or even falsified data do exist and wrong interpretation of even reliable results adds to confusion in scientific and public discussions. In view of the severe impact of environmental radioactivity data on the public opinion and on society in general, proof of quality and reliability is a most important issue and a basic requirement for measurements of environmental radioactivity. This requires a sophisticated system of quality assurance and control which shall shortly be discussed here.

A first requirement is that the purpose of a measurement and thereby the measurand are unambiguously defined. It is a difference whether the measurand is the activity concentration of particular sample or whether it is the activity concentration of a particular environmental material. While in the first case the measurand can be assumed to be well defined in the second

case the question whether a sample or a set of sample is representative for the material in question is a difficult problem and usually the measurand in this case is not well defined.

The quality of a measurement starts with sampling strategy, sampling methods and their documentation. The choice of the measurand and the sampling strategy are influenced by the purpose of the measurements, see Gilbert (1987) for some discussion of the general problems of sampling. It can even be the case that it is not possible to define the measurand exactly and that only distributions of results of measurements can be used to attack the problem in question. As an example, the frequently observed log-normal distribution of activity concentrations in environmental materials must not necessarily be a consequence of unreliable measurements. It represents in the optimum case of reliable measurements of individual samples the distribution of the true values of the measurands in the samples of an environmental compartment. This effect is *e.g.* caused by input and output probabilities for a compartment which are linearly normal distributed. On the other hand, such a distribution can be caused by insufficient control of analytical blanks and by systematic influences on the measurements which are not properly corrected for. Therefore, to judge about the question whether a distribution of results is an experimental artifact or whether it represents the natural conditions can only be decided by a thorough investigation and determination of the uncertainties of the measurement.

With the ISO Guide for the Expression of Uncertainty in Measurement (ISO, 1995) an internationally accepted unified approach to the determination of experimental uncertainties does exist. In contrast to still frequent practice it requires to take into consideration all sources of uncertainty. In particular, it distinguishes between Type A and Type B uncertainties. Type A uncertainties are those which can be evaluated by the experimentalist by repeated or counting measurements and which can be expressed as standard deviations. Type B uncertainties are those which cannot be evaluated by the experimentalist himself but which he can obtain them only from other sources. Type B uncertainties are, for instance, reported in literature or stated in certificates, *e.g.* for calibration standards and nuclear decay data used. While Type A uncertainties can nicely be evaluated by conventional statistics, Type B ones cannot. Using, however, Bayesian statistics also Type B uncertainties can be expressed as standard deviations and can be propagated to combined standard uncertainties associated with a measurement result as the best estimate of the true value of the measurand (Weise and Wöger, 1993, 1999). In this case, also a probability distribution for the true value of the measurand can be given which makes use of any existing information and which can be improved if further information emerges. It is a *conditio sine qua non* that for any measurement an analysis of the experimental uncertainties has to be performed. A result without a statement of the combined standard uncertainty associated with it and with a list of all considered sources of uncertainties is no result and worthless. Considering, however, the determination of the human internal radiation exposure there are still some unresolved issues since there are no uncertainties of dose factors available. Here further investigations are urgently needed.

If experimental uncertainties are known according to the ISO Guide also the problem of an unambiguous definition of decision thresholds, detection limits and limits of the confidence interval (named characteristic limits in short) can be made (Weise, 1998). In an upcoming ISO standard (ISO, 2000a) the determination of characteristic limits will be internationally laid down. A national German standard on this issue is already published (DIN, 2000).

The approach to the determination of characteristic limits is general as the approach for the evaluation of uncertainties. It does not only apply to radioactivity measurements but can easily extended to any analytical measurement where a measurand of quantity in a sample has to

be measured against a background or blank of the general environment (Michel, 2000). The determination of a net concentration of a chemical element against the analytical blank is an example here.

Generally, the determination of characteristic limits according to the cited standards is straight forward and only in some cases as in γ -spectrometry some more sophisticated mathematical methods have to be applied (Weise and Michel, 1995).

The commitment to a unified approach for the determination of characteristic limits is also of another practical importance. Since required detection limits are used in many countries to specify requirements for methods used for environmental radioactivity measurements, an unambiguous definition of detection limits is necessary. The reporting levels used by the European Union (European Commission, 1996b) in practice have the character of required detection limits since results of measurements are discarded for which the detection limit is not below the reporting level. There is, however, some confusion that the respective report uses the term detection limit but it does not specify how the detection limits have to be calculated and does not give the probabilities of the errors of 1st and 2nd kind on which the detection limit has to be based. It becomes even worse if one takes the example of the Annex of the EU Drinking Water Guideline (European Commission, 1998, 2000b) where “*required detection limits*” are given for radioactivity measurements. Again there is no meaningful specification of the definition of detection limits and with regard to other analytical investigations of the water even contradictory statements are made. Also in the general scientific literature widely inadequate use is made of characteristic limits and insufficient information is frequently given about the conditions under which these limits were determined. Therefore, a standardized use of characteristic limits is urgently needed as a part of setting unified requirement for measurements and for an unambiguous report of such limits for a measurement. With the upcoming ISO 11929-7 (ISO, 2000a) or with DIN 25482-10 (DIN, 2000) such a standardized approach is possible and a requirement for any measurement of environmental radioactivity.

An experimental and analytical problem generally arises from the fact that environmental radioactivity has always to be measured against an environmental background and/or an analytical blank. While it is standard practice to correct for counting or spectrometric background in the measurements themselves, the interference by analytical blanks is frequently still neglected. One can make the statement that any measurement claiming an enhanced abundance of a radionuclide or element in a sample is worthless if no information on the analytical and/or environmental blank is given. Though the use of clean rooms and high-purity reagents may considerably reduce analytical blanks the demand is with the experimentalist to demonstrate and to report in a traceable form the actual blanks. An impressive example for blank problems is a recent round robin exercise for the determination of ^{129}I in environmental materials (Roberts *et al.*, 1997, 2000). While the measurements of ^{129}I in readily prepared AgI samples by AMS proved to agree within reasonable limits, the results for the entire analysis of ^{129}I in environmental samples scattered by orders of magnitude. This disagreement could be pinpointed to result from differences in the analytical blanks of the various laboratories. For ^{129}I and ^{36}Cl differences of orders of magnitude in the analytical blanks can easily occur, see *e.g.* (Schmidt *et al.*, 1998) and only repeated and consequent blank analyses can provide a basis for accurate results.

A general guaranty for accuracy can, however, not be given. To demonstrate accuracy and reliability, repeated analyses of reference materials and frequent participation in round robin exercises or interlaboratory comparisons are essential. Large efforts have been undertaken to establish reference materials. However, in spite of the intensive work done for instance by the

IAEA and many national or other international institutions, for many materials reference materials are still lacking and the further development of new materials can only be highly encouraged.

In view of the general globalization process, which affects also the discussions of environmental radioactivity, interlaboratory exercises on an international level have to be preferred to national ones. It is, however, important that the same standardized and evaluated methods are used for the analyses of samples from such exercises and for normal ones of the daily work in order to assure that the same claim of accuracy can be made for all results obtained by a laboratory. In this context, quality assurance and control on the basis of national and preferably of international standards becomes essential. Only by use of strict analytical protocols and completely traceable performance of measurements it is probable to guarantee quality.

Evaluated procedures for measurements of environmental radioactivity do exist in many countries (AFNOR, 2000; AKU, 1999; BMU, 1998; DoE, 1979), but only few of them are standardized on a national or international level. Thus, ISO TC 85 SC 2 has set the goal of establishing such standards in the future and work has begun to develop respective standards starting with the measurements of radionuclides in soil (ISO, 2000b). This standard will not only deal with the radiochemical and analytical aspects of soils, but also with sampling strategies and methods which just recently have been investigated by international collaborations (Anonymous, 2001). It will still take some time until internationally accepted standardized procedures will be available. But, such standards are urgently needed to qualify and certify laboratories performing measurements of environmental radioactivity.

It is a general trend here that certification of laboratories will be required in the future if measurements are performed on a commercial basis or if legally required measurements have to be performed. Peer review of scientific investigations and publications has strictly to investigate whether adequate measures of quality assurance and control have been taken if no certification for a research group does exist. But, even with all possible measures of quality assurance and control, the struggle for accuracy and reliability will never end.

As a further point with respect to quality it has to be emphasized that any judgment about anomalous conditions, i.e. enhanced radioactivity in the environment, requires as a basis detailed knowledge about what is normal. This knowledge is not always available. As an example, the existing data about naturally occurring radionuclides in many cases are strongly biased due to the fact that samples were taken more frequently in regions where geologically enhanced radioactivity was expected. There is a considerable lack of data in geologic low-activity areas for nearly all environmental materials. This lack of knowledge and the consequent bias makes reasonable judgement about NORM and its influences on the human radiation exposure severely difficult.

Finally, the documentation of the results is an important facet of the quality of a measurement. It is, moreover, a basic requirement for traceability of a result. It has to be as complete as possible describing the sampling aspects, the analytical methods, the measures of quality assurance and control used, the standards on which the result is based and all information including the nuclear data used for evaluation. It has to state the result and its combined standard uncertainty, the characteristic limits and the way they are computed including the assumed probabilities of the errors of the 1st and 2nd kind and the probability considered for calculating the confidence limits. The results of blank analyses and of reference materials also have to be included into the documentation.

Given the enormous number of measurements of environmental radioactivity all over the world, adequate documentation becomes a major problem if just printing media are considered because of the tremendous amount of information needed to judge about a measurement result. Here, new methods of electronic documentation and publication are urgently needed to inform the scientific world as well as the general public.

5 Environmental Radioactivity and the Public

In the public perception, radioactivity, in general, and radioactivity in the environment, in particular, is directly connected with the risk of stochastic effects of ionizing radiation and discussed with emotion and belief rather than rationally. Without discussing the reasons for the irrational public perceptions of risk and radioactivity (Pretre, 1992, 2000), any attempt must be undertaken to rationalize the way societies treat these issues. The complexity of this problem is that it contains a sequence of questions which only partially can be solved by the methods of natural sciences. It starts with the question: What can we know about the abundances of radionuclides in the different environmental materials and what is the resulting radiation exposure? It continues with: What is the risk associated with this radiation exposure? and it ends with the questions: Is the risk acceptable for society, for the groups at risk and for the individual? and What shall society and the individual do?

When dealing with these complex issues in the public, measurements of environmental radioactivity provide the fundamental data, on which the evaluation of the resulting radiation exposure is based. Therefore, these data must be of outstanding quality and beyond any doubt because, otherwise, there is no scientific basis for any subsequent public discussion. The fact, that the determination of the risks associated with an exposure is not finally settled adds already enough confusion to the public discussion. Still existing lack of knowledge about the induction and mechanisms of stochastic effects and the fact that the risks of radiation exposure have to be evaluated on the basis of incomplete information as *e.g.* the future frequency of cancer and leukemia in the still living survivors of the Hiroshima and Nagasaki explosions (Nagataki, 2000).

To improve the present unsatisfactory situation it will not be sufficient to keep the knowledge of the abundances of radionuclides in the environment and of the resulting exposure in the scientific community. All existing information must be broadly disseminated and has to be freely and easily available in forms which are understandable for the individual target groups which cover, besides scientists, governments, regulators bodies, politicians and decision makers, media, environmental activists, political parties and the silent majority of the population.

The present habits of publishing only in scientific journals and of preparing excellent national and international reports on these issues are not enough. They must be accompanied by availability of all required information in the electronic media with complete transparency. To this end a hierarchy of informative systems has to be established nationally and internationally which can be realized by the use of the internet. The hierarchical structure will result from the degree of in-depth information expected by the respective user group. It must range from a level of general information to the possibility of accessing the real data. Such a system has to take special care of the needs of children in schools and students in academic education. While present days adults widely are influenced by opinions and beliefs resulting from the threads of the cold war, more rational perception of radioactivity and radiation exposure will widely rely whether respective information can be disseminated to young people.

In public discussions of an actual problem one has to accept that measurements of the really existing radionuclide concentrations will be required even if those concentrations are beyond

any radiological relevance. It has, however, to be made clear to the public, that the timeliness of respective results cannot be always guaranteed since such measurements occur on time-scales which may be long compared to those of discussions in a society. The invention of such in-depth measurements in the coarse network used by the EU in surveillance of environmental radioactivity (European Commission, 1996b) is a good example for what has to be done if an actual issue is in the public discussion. Moreover, the results obtained by the Nord-Cotentin Radioecology Group (1999, 2000) in the discussion of the exposure and the induced risk from the emissions from the La Hague reprocessing plant are an outstanding example here.

Another problem arises from the way actual exposures are evaluated and presented to the public. The evaluation of the human exposure is the ultimate goal of environmental radioactivity measurement. The EURATOM basic safety guidelines (European Commission, 1996a) require the surveillance of the human radiation exposure, in general, and the detailed evaluation for groups with elevated exposure. They further demand that the exposure has to be determined as realistic as possible. There is, however, a wide-spread confusion due to the fact that conservative approaches are frequently used to model exposures the results of which may exceed a realistic exposure estimate by orders of magnitude. While these conservative approaches are fully acceptable and justified for planning purposes they must not be used to estimate the actual exposure in a given situation where measured data exist and more reliable and realistic dose estimates can be made. In the future information policy clear distinctions between these case of exposure estimates have to be made.

Finally, an important issue for the public discussion is the consistent treatment of exposures due to natural and man-made radioactivity. There is a discrepancy between the perception of the large variability of the natural radiation exposure, on the one hand, and of the usually very small contributions of man-made radionuclides, on the other. Even in accidental situations or large scale contaminations such discrepant judgements can be observed. On the basis of the requirements of *e.g.* the EURATOM basic safety standards (European Commission, 1996a) and of an increasing awareness of the scientific and regulatory communities it will be an important issue for future considerations to struggle for a consistent system of radiation protection when dealing with natural and man-made radioactivity.

6 Future Trends and New Frontiers

Summarizing, future trends of measuring environmental radioactivity can be recognized with respect to the development of the measurements themselves and of the analytical practices, with respect to their fields of applications and with respect to the dissemination of the results.

In parallel to the further refinement of existing methods of nuclear radiation spectrometry, the future developments will comprise increasing applications of mass spectrometric measurements. The analytical techniques will be further improved by especially tailored chemical reagents for extraction of radionuclides from environmental materials and by the development of automated chemical systems for separation of a large number of radionuclides from one sample. Finally, the laboratory practice will be dominated in the future by systems of quality assurance and control. Certification of laboratories and complete traceability of the results will be state-of-the-art without which no measurements will be acceptable.

The purposes of environmental radioactivity measurements will improve from documentation of man-made changes of the environment to measurements dedicated to the achievement of a sustainable development. This requires

- surveillance of (further minimized) emissions from nuclear installation with the goal of to attain negligible emissions,
- guidance of measures for remediation of contaminated territories and sites and for a safe disposal of radioactive waste, and
- quantification of the long-term impact of man on the radiation exposure of man and other parts of the biosphere in addition to the evaluation of the short-term impact of man.

Moreover, there will be an increasing importance of measuring natural radioactivity and potentially enhanced exposures to these nuclides from non-nuclear technologies. Finally, measurements of environmental radioactivity will be increasingly used to exploit the information about environmental processes in which long-lived radionuclides act as tracers and document both natural and man-made environmental change.

Last but not least, the future will see a widely changed attitude with respect to the dissemination of the results and the information of the public which besides quality of the measurement results is the key issue for a rational perception of environmental radioactivity in society.

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