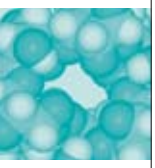
V. STANDARDISATION ASPECTS

The pre-existence and authorised or accidental additions of radioactive substances in the environment justify a demand for the expert assessment of the radiological state of the environment (radioecology) and the evaluation of impact on human populations (radioprotection). This demand is voiced by international, national or regional official bodies and nuclear plant operators, but also by the media and by local public associations. In addition, awareness of the importance of environmental protection in a context of sustainable development has resulted in a demand for reference methods to enable users of nuclear technology to evaluate the environmental effects of their activities. Thus even though the levels of radioactivity currently expected in the environment are low, measurement results are needed to establish the impact of changing practice and of nuclear accidents. The results of these measurements can be used to determine local radiological status or their long-term time course or to explain and quantify the mechanisms of transfer of radionuclides between different parts of the environment. They may be compared with the results of calculations obtained by mathematical simulation, in order to validate theoretical impact and hazard evaluations. The credibility of expert assessment in the "nuclear and environment" area partly relies therefore on the quality and reliability of strategies for the collection of environmental samples, their laboratory processing and the assay of the radionuclides they contain. To quote a report of the Academy of Science, *"measurement issues are central in environmental studies"*. This is especially true in the nuclear field and certainly more so than elsewhere.



There is a pressing need for universally accepted references governing the description of the radiological state of the environment, that will meet the demands of official bodies, industry and public opinion. The development of recognised standards in this area is thus necessary.

Standardising the measurement of radionuclides in the environment

There is a current trend towards the internationalisation of the inspection and monitoring of industrial activities through the adoption of international treaties and agreements for the evaluation of the impact of chronic or accidental cross-frontier contamination. For national nuclear industries and their relationship with the environment, this extension of partners to include those in other states requires the results of the measurement(1) of **activities**, presented by a country to characterise effluents released into the environment, or environmental samples, to be reliable and reproducible so that they are acceptable by all the states potentially concerned by a regional contamination.

The methods and means of measurement have therefore to keep pace with the developments and modifications of practice linked to the evolution of nuclear applications, which has resulted in a progressive reduction in the release of **radionuclides** into the environment. Today, measuring the levels of activity of artificial radionuclides in the different aquatic and terrestrial media over the greater part of France means measuring trace amounts(2) at the very most. In all cases the methods selected have to meet the requirements of **ISO** (International standardization organization) standards for test laboratories in order to guarantee the reliability of measurement results that is necessary to ensure the credibility of research findings, expert reports and regulatory inspections.

Here we briefly present the work being done on standardisation and the consistency rules necessary for the measurement of radionuclides in environmental compartments. The standardisation documents in force are also reviewed.

Measurement of radionuclides in the environment

Expert reports on naturally-occurring radionuclides are usually concerned by the remobilisation of **elements**, resulting in small additional amounts of activity relative to natural levels. For artificial radionuclides, inspections have shown that their activities are generally very much lower than those of the other naturally-occurring **radioactive** constituents of the environment. In addition, contamination from reagents, equipment or the laboratory environment can lead to overestimated values. Losses of the **analyte** being measured, during radiochemical processing operations, for example, are more critical at low concentrations or when small discrepancies in activity are being measured, and can make it impossible to detect the analyte or discern a variation in its activity. Lastly, interference between the constituents of the sample and the detection system can lead to under- or overestimated values, making it necessary to add purification steps or to use more selective detectors.

To respond to these constraints, the metrologist therefore has to lower detection thresholds and increase the precision and accuracy of the measurements. In this context, guaranteeing the reliability of the results obtained is considered as a crucial issue for the analysis of radionuclides in the environment.

The only way to achieve this guarantee is through the setting up of a strict quality assurance system that complies with good laboratory practice (OECD, 1992) and with the requirements stated in the principles of quality assurance, codified in the ISO 9001 series of documents for certification, and in the ISO standard 17025 for accreditation.

Organisation of standardisation in the nuclear field

The Nuclear Facilities Standardisation Office (BNEN) and the French Standardisation Association (Afnor) are the bodies concerned with standardisation in metrology in France. Internationally, the European Standardisation Committee (CEN) and ISO are engaged in the definition of strict metrologoical practice for the measurement of radionuclide activities, among other parameters, in order to ensure that results obtained in the different Member States are comparable.

(1) As employed here, *measurement* is the ISO standard term that covers all the operations that aim to determine the value of a quantity (from the international vocabulary for fundamental and general terms of metrology (ISO). In this case, the measurement serves to determine the activity of a **radionuclide** expressed in **becquerels** (Bq).

(2) Trace amounts: many analysts use this term to describe amounts less than one part per million (ppm); others use it when dealing with analytes present at concentrations for which it is technically difficult to obtain validated results.

CEA/Valduc

CEA/Valduc

Measurement of physicochemical parameters of water in samples taken from the environment of a CEA centre. Inset: filter record corresponding to a weekly air sample before dispatch to nuclear analysis laboratory.

In France, the BNEN, certified on June 26 1990 by the Ministry for Industry and Planning, is responsible for overseeing the French commissions and associated working groups (Figure p. 98), in the areas of protection against **ionising radiation** (Commission M60-1), nuclear **fuel** cycle technology (M60-2) and measurement of radioactivity in the environment (M60-3). This last commission was set up in 1992 by order of the Ministry for Industry following disputes over the results of measurements obtained by different laboratories. The Ministry asked Afnor to make proposals for standardising work on the measurement of radioactivity in the environment. A standardising approach seemed most likely to forestall disagreements that might otherwise arise among different public and private bodies that found themselves issuing significantly different activity results from apparently identical samples.

At CEA, the Cetama(3) is in charge of taking action to improve the quality of the results of measurements made in its laboratories. Fourteen working groups, including Working Group 31 (Analysis of radionuclides in the environment), organise intercomparison circuits, and validate and draft measurement protocols. Some of these methods are proposed to the BNEN working groups for evaluation by the entire metrological community, and sometimes submitted to Afnor as projects for standards.

To meet the objectives of Afnor, Commission M60-3 was organised in four working groups reflecting the main environmental media: air, water, soil and bioindicators. This structure also addresses the sections of Programme 135 of the French accreditation committee (Cofrac). A fifth working group on food was added to meet the requirements of Cofrac Programme 99-4.

This commission is responsible for standardising the methods of measurement of radioactivity in the environment, outside the fields of medicine, **radioprotection** of workers, and contamination. Its commission is composite, its members being directly responsible for monitoring the environment near EDF, Cogema and CEA facilities, or in charge of inspection (DGSNR) or assessment (IRSN), or are members of an association for environmental protection such as Criirad. For those responsible for monitoring, acceptance as a formal standard of a measurement method used in the laboratories is a recognition by all the partners in the area of the quality of the work conducted in these units.

Since its creation, the work of this commission has resulted in the drafting of more than 40 Afnor standards concerning methods of taking environmental samples and measurement of radionuclides. The drafting of some ten projects for additional standards is in progress.

Standardisation documents

Radioecology studies and health impact assessments are based on the results of measurements made on environmental and food samples initially taken using what are known as generic methods, which are described in international publications (**IAEA**, **WHO**). When test laboratories voiced their need for specific formal standards, new documents were produced by Commission M60-3 which, as we shall

(3) The Cetama (Commission for the specification of analytical methods) is responsible for taking action to improve the quality of the results of measurements made in the laboratories of CEA.

CEA/DAM/IdF

The choice of samples in the field has to be representative of the situations being studied and requires complementary knowledge of pedological and hydrogeological parameters, studied here from the data supplied by water probes and a weather station.

see below, took position among the already published Afnor and ISO standards, occasionally among those of the American ASTM (ASTM International, previously American Society for Testing and Materials) and of the International Electrotechnical Commission (IEC), more specifically concerned with instrumentation and equipment based on electronic components.

Water quality

The standardising work carried out by the Water Group is intended for the laboratories responsible for measuring the radioactivity present in drinking water and untreated water with a low mineral content. The efforts of Afnor, and later ISO, in the water quality field, has been important and has culminated in the publication of an almost complete set of standardisation documents applicable to radioecological assessment and research. Documents complementary to those of ISO have been published by Commission M60-3 (Table), in particular for the determination of gross **alpha** (NF M60-801) and **beta** (NF M60-800) **activity** indices in water with low mineral content, the updating of the standard for tritium measurement in water (NF M60-802-1) and the standard for carbon-14 measurement (NF M60-802-2), and then for both radionuclides simultaneously (NF M60-802-3). The monitoring of tritium levels in drinking water has been the subject of special attention in Europe (CEC, 1998), because of current release from nuclear power plants, which is a non-negligible local source of environmental tritium. The **radiotoxicity** of tritium is considered weak, but the volumic activity of tritium in the environment is monitored so as to be able to track its circulation in the hydrosphere and **biosphere** through to drinking water. A standard has been published for the measurement of the activity of radium-226 in water (NF M60-803).

After the revision of Directive 80/778/CEC concerning the quality of drinking water, which culminated in Directive 98/83 concerning the quality of water for human consumption, the existence of Afnor standards on this subject is important for the technical discussions that will be taking place at the European level.

Air quality

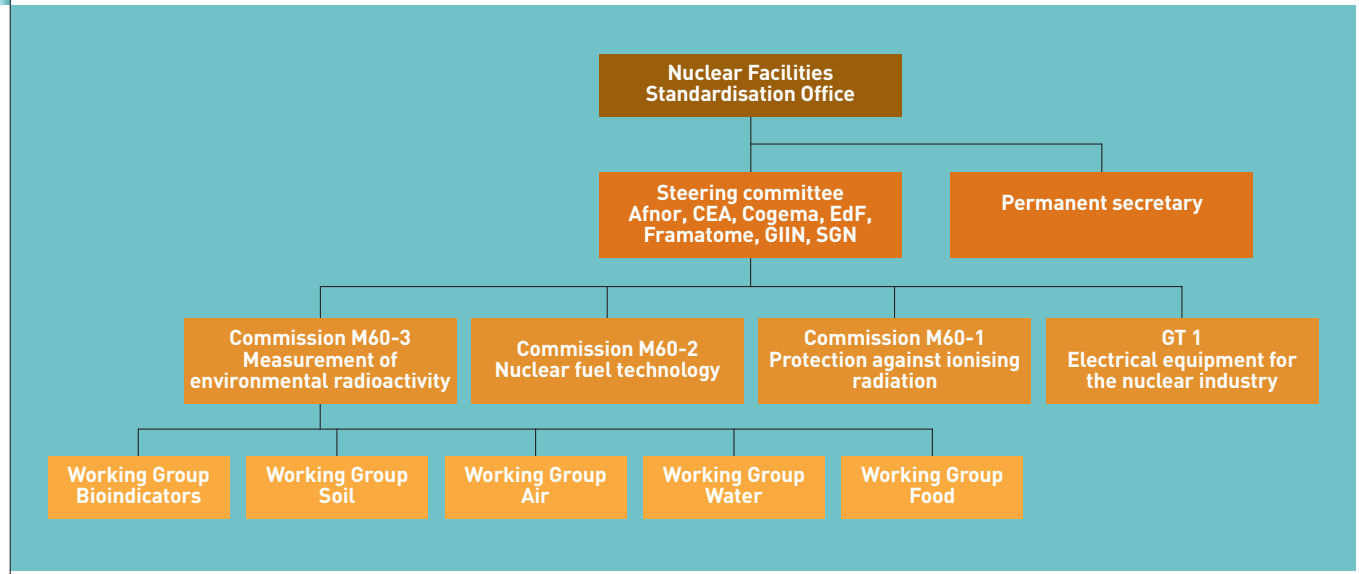
The standardising work of Afnor and ISO in the field of air quality has mainly concerned the general principles for the sampling of radionuclides present in the air, with heavy emphasis on documents concerning those associated with the particulate fraction. The CEI has issued standards on the measurement of tritium and on the instrumentation for the measurement of **radon** and its decay products. Commission M60-3 is accordingly engaged in reviewing the standard for the volumic assay of atmospheric tritium and the drawing up of complementary standards for the sampling of **aerosols** (M60-760), the activity of soil deposits (M60-770) and the measurement of **halogens**. Efforts have been focused in particular on the measurement of radon, a naturally-occurring radioactive gas produced by the **decay** of uranium and radium in the ground. Radon reaches significant concentrations in the air in regions with granite and volcanic substrata, and can also be released from building materials.
Radon and its solid decay products are **inhaled** and irradiate the lungs. Accordingly, to meet the requirements of the joint DGS and DGUHC circular No. 99/46 of January 27 1999, stating the action to be taken to control the radon hazard in buildings, a special effort has been made as regards the different methods of measurement of radon-222 and its decay products. Eight standards have been published on this subject (radon-222 series).
On the topic of radon, we also note that the French translation of the ISO 13466 standard has been validated by the members of Commission M60-3. In addition, in the framework of the standardisation work of the CEI, members of the commission participate in Working Group 45 B.

Soil quality

The Afnor and ISO standards for soil quality concern work on the general principles for the sampling and preparing of samples before the detection of radionuclides. They form part of the methods for site studies, in particular those used in radioecology. Commission M 60-3 therefore re-examined the existing standardisation documents to adapt them to the questions arising from expert assessment and research requiring the measurement of radionuclides, in particular for indices of gross alpha radioactivity, **gamma emitters**, strontium-90 and plutonium. The same approach was taken by the ASTM.
The soil working group thus drew up eight Afnor standards (M60-790 series). It set out to respond to requirements of the bodies responsible for determining the main radionuclides present in soil, whether in routine monitoring, expert assessment or initial characterisation of a site, studies of vertical distribution of radionuclides in soils, or studies of surface radioactivity of soils, with a view to studying

reference	title of standard
Water quality	
NF M60-800	measurement of strontium-90 and yttrium-90 equivalent global beta radioactivity in water with low mineral content
NF M60-801	measurement of plutonium-239 equivalent alpha radioactivity in water with low mineral content
NF M60-802-1	measurement of beta emitters by liquid scintillation: special case of tritium
NF M60-802-2	measurement of beta emitters by liquid scintillation: special case of carbon-14
NF M60-802-3	measurement of beta emitters by liquid scintillation: special case of simultaneous presence of tritium and carbon-14
NF M60-803	measurement of radium-226 activity in water
NF M60-804-1	measurement of the activity of transuranics (Pu, Am, Cm, Np) by alpha spectrometry in water - general
NF M60-805-1	measurement of the concentration of uranium in water by fluorimetry
NF M60-805-2	measurement of the concentration of uranium in water by atomic emission spectrometry with induction coupled plasma
NF M60-805-5	measurement of the activity of uranium in water by alpha spectrometry
Air quality	
NF M60-312	determination of the volumic activity of tritium in the atmospheric environment
NF M60-760	determination by liquid scintillation of the volumic activity of atmospheric tritium sampled by the method of bubbling air through water
NF M60-761	determination of the volumic activity of radioactive halogens in the atmospheric environment
NF M60-763	radon-222 and its short-lived decay products in the atmospheric environment: origins and measurement methods
NF M60 764	radon-222: methods for the integrated measurement of the potential volumic alpha energy of the short-lived decay products of radon in the atmospheric environment
NF M60-765	radon-222: methods for the point measurement of the potential volumic alpha energy of the short-lived decay products of radon in the atmospheric environment.
NF M60-766	radon-222: methods for the integrated measurement of the mean volumic alpha energy of radon in the atmospheric environment with passive sampling and delayed analysis.
NF M60-767	radon-222: methods of continuous measurement of the volumic activity of radon in the atmospheric environment
NF M60-768	radon-222: methods of estimation of surface exhalation flux by the accumulation method
NF M60-769	radon-222: method of point measurement of the volumic activity of radon in the atmospheric environment
NF M60-770	collection of atmospheric deposits on soil and determination of the activity of atmospheric deposits on soil
NF M60-771	radon-222 in buildings: methods applied to screening and complementary investigations
Soil quality	
NF M60-790-1	part 1: general presentation and definitions
NF M60-790-2	part 2: guide for the selection of sample collection areas, sampling, and transport and conservation of soil samples
NF M60-790-3	part 3: method for the pre-treatment of soil samples
NF M60-790-4	part 4: method for the dissolution of soil samples
NF M60-790-5	part 5: method for the measurement of ^{239}Pu equivalent global alpha radioactivity index and ^{90}Sr and ^{90}Y equivalent global beta radioactivity index at equilibrium in soil samples
NF M60-790-6	part 6: method for the measurement of gamma emitter activity in soil samples
NF M60-790-7	part 7: methods for the measurement of strontium-90 in soil samples
NF M60-790-8	part 8: methods for the measurement of isotopes of plutonium (plutonium-238 and plutonium-239+240) in soil samples
Bioindicators	
NF M60-780-0	general principles
NF M60-780-1	part 1: general guide for drawing up sampling programmes
NF M60-780-2	part 2: general guide on sampling methods
NF M60-780-3	part 3: general guide for the conservation and handling of samples
NF M60-780-4	part 4: general guide for sample preparation
NF M60-780-5	part 5: general guide for sampling biological indicators in terrestrial media
NF M60-780-6	part 6: general guide for sampling biological indicators in fresh-water media
NF M60-780-7	part 7: general guide for sampling biological indicators in marine media
NF M60-780-8	part 8: glossary
Food	
NF V 03 009-1	measurement of radioactivity in foods - guide to sampling, transport and conservation of foods - obtaining a laboratory sample
NF V 03 009-2	measurement of radioactivity in foods - guide to the preparation of food samples – obtaining a test sample

Table.
Titles of standards drafted by the working groups of Commission M60-3 (Measurement of radioactivity in the environment) and published by Afnor.



Organisation of the Nuclear Facilities Standardisation Office in which four working groups deal specifically with the methods for measuring radionuclides in four major types of environmental medium: air, water, soil and bioindicator organisms.

atmospheric fallout or re-suspension. These standards are used to produce reliable data necessary for simplified detailed risk assessment justifying the cleaning up of old industrial sites, the soils of which may have been contaminated by radionuclides (IRSN guide).
The first standard (NF M60-790-1) sets the general framework and the successive steps. The second standard is a guide for the selection of sampling points and areas, and the transport and storage of soil samples (NF M60-790-2). The other six standards cover methods of work-up and measurement of soil samples in the laboratory. Standards 790-3 and 790-4 concern respectively the drying, crushing, sieving and homogenisation of the samples, and the dissolution of the radionuclides to be measured, a necessary step in the selective testing for pure beta emitters (e.g., strontium-90) and alpha emitters (**isotopes** 238, 239 and 240 of plutonium). The measurement of radioactivity indices of global alpha and global beta emitters, which provides a first evaluation of the dispersal of activity levels, is covered by the standard NF M60-790-5. The measurement of gamma emitting radionuclides, in particular the main products of the natural **decay chains** of uranium and thorium, and the artificial radionuclides including cæsium-137, which is present in all surface soil samples (fallout from atmospheric nuclear weapon tests and the Chernobyl accident) is dealt with in the standard NF M60-790-6. Parts 8 and 9 of this series respectively cover the measurement of strontium-90 and the alpha-emitting isotopes of plutonium (plutonium-238, -239 and -240). They involve complex operating procedures justified by the **potential radiotoxicity** of these elements. This work completes that of ISO on marine and aquatic sediments.

Bioindicators

Living organisms, in particular bioindicators characterised by a high capacity for accumulation of industrial contaminants, including radionuclides, can be used to evaluate qualitative and quantitative trends in the contamination of natural media. Bioindicators are particularly useful in that they integrate all the forms of radionuclide release liable to have an impact on the environment.
However, if no precautions are taken in the storage of samples, biological indicators are liable to be degraded between the time of sampling and that of laboratory analysis. The conservation of samples during transport and while they are being stored in the laboratory pending analysis, has to ensure that the concentrations determined are representative of those in the original sample, in particular for the main elements making up living matter.
The specification of a Bioindicators Group in Commission M60-3, which does not appear in the other standardising structures, brought this group close to existing Afnor and ISO standards in the areas of water quality, agricultural produce and food.
Standardisation work culminated in the drafting of general guides on the precautions to be taken for the sampling, storage and handling of bioindicator material before its processing in the laboratory (M60-780-1, -2, -3, -4). A second series of guides concerning the sampling of biological indicators specific to different terrestrial, fresh-water and marine media (M60-780-6, -7, -8) has also been published.

Food

For the food component, for France, the work planned by the BNEN working group was presented to the strategic orientation committee (COS) for the agrifood sector. The programme led to the drafting of two standards that complete those produced under the ægis of this COS.

Recognised quality of French projects

The potential contamination of the environment by radioactive matter is a regular subject of public controversy. This being so, it is easy to appreciate the importance of being able precisely to identify and quantify a source of environmental contamination, to discern the respective contributions of different

sources, and (or) to be able to rule some out. In these situations the quality and reliability of sampling strategies, the taking of samples and the results of measurements have to be consensually approved by all the interested parties so that any ensuing discussion remains focused on interpreting the results and not on challenging their validity. Measurement must therefore be supported by a method for exploring the medium and recognised metrological procedures referenced if possible in standards that are themselves the result of a consensus among the different partners: industry, regulators, and official or independent experts. By providing an unequivocal technical response based on samples that are representative of the environmental levels the radioactive content of which is to be measured by a certified or accredited test laboratory, an impact assessment derived from the radioactivity data will benefit in term of credibility.

The development of standardisation in the field of radioactivity measurement, in particular in France, thus attests to a desire on the part of all those involved to pool their expertise and improve clarity in their area. This aim of clarity is shared by the public authorities. The availability of these standards means they can be used as a basis for decision-making. This is an important aspect, and the participation of CEA specialists is indispensable to voice the viewpoint of both a representative of the nuclear industry and an expert body in this field. The prime objective of the members of Commission M60-3 is thus to complete a corpus of standards for the measurement of radionuclides present in the different **compartments** of the environment (air, water, soil and biological indicators). This collective work, with the participation of CEA experts, besides responding to the initial institutional demands from the ministries in charge of healthcare, ecology and industry, and from local authorities, contributes to a better appraisal of the impact of the nuclear industry and its role in improving the quality of our environment in a context of sustainable development. We note the sometimes innovative character of the work carried out by this commission, especially that on bioindicators. This corpus of standards will thus form a set of references with no equivalent internationally. The decision was thus taken, in particular by BNEN and Afnor, to propose the internationalisation of these standards, i.e., their recognition by ISO. In 1999, the set of standards for the measurement of radionuclides in soils was submitted to the vote of ISO as a new subject for study to be dealt with in ISO/TC 85/SC 2/WG 17. The ISO member countries accepted this project, thereby recognising the quality of the work. The first review of the French projects was carried out in May 2000 at the following meeting of this working group. The projects of revised standards are now circulating at the level of subcommittee 2. To pursue this process the next submission to ISO is to cover the standards concerning the measurement of radionuclides in water. Overall, we stress the high level of international interest shown in this work, which further demonstrates their importance and utility in addressing issues that are now of international concern.

DarkhorseStudio/Cogema

Sample of fresh water taken at the site of Cluff Lake, Saskatchewan (Canada) by a technician from Cogema.

> Dominique Calmet*
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CEA - DAM-Ile de France Centre
*Chairman of BNEN Commission M60-3 and ISO project leader for measurement of radioactivity in the environment.

A Natural and artificial radioactivity

Everything on the earth's surface has always been exposed to the action of ionising radiation from natural sources. Natural radiation, which accounts for 85.5% of total radioactivity (natural plus artificial), is made up of 71% telluric radiation and about 14.5% cosmic radiation. The radionuclides formed by the interaction of **cosmic rays** arriving from stars, and especially the Sun, with the nuclei of elements present in the atmosphere (oxygen and nitrogen) are, in decreasing order of dose (Box F, *From rays to dose*) received by the population, carbon-14, beryllium-7, sodium-22 and tritium (hydrogen-3). The last two are responsible for only very low doses.

Carbon-14, with a half life of **5,730 years**, is found in the human body. Its activity per unit mass of carbon has varied over time: it has diminished as carbon dioxide emissions from the combustion of fossil fuels have risen, then was increased by atmospheric nuclear weapon tests.

Beryllium-7, with a half life of **53.6 days**, falls onto the leaf surfaces of plants and enters the body by ingestion (Box B, *Human exposure routes*). About 50 Bq (becquerels) per person per year of beryllium-7 are ingested.

The main or **"primordial" radionuclides** are potassium-40, uranium-238 and thorium-232. Along with their radioactive decay products, these elements are present in rocks and soil and are therefore found in many building materials. Their concentrations are generally very low, but vary according to the nature of the mineral. The gamma radiation emitted by these radionuclides forms the **telluric radiation**, which is responsible for the external exposure of the body. The primordial radionuclides and many of their long-lived descendants are also found in trace amounts in drinking water and plants: this results in an internal exposure by ingestion, plus an additional low exposure by inhalation of airborne suspended dust particles.

Potassium-40 is a beta and gamma emitter with a half life of **1.2 thousand million years**, and has no radioactive descendants. This radioactive isotope makes up 0.0118% of all natural potassium, and enters the body by ingestion. The mass of natural potassium in the human body is independent of the quantity ingested.

Uranium-238 is an alpha emitter with a half life of **4.47 thousand million years**. It has thirteen main alpha-, beta- and gamma-emitting radioactive descendants, including **radon-222** (**3.82 days**) and **uranium-234** (**0.246 million years**). Uranium-238 and its two descendants **thorium-234** (**24.1 days**) and **protactinium-234m**(1) (**1.18 min**), and **uranium-234** are essentially incorporated by ingestion and are mainly concentrated in the bones and kidneys.

Thorium-230, derived from uranium-234, is an alpha emitter with a period of **80,000 years**. It is an osteotrope, but enters the body mainly by the pulmonary route (inhalation). **Radium-226**, a descendant of thorium-230, is an alpha emitter with a half life of **1,600 years**. It is also an osteotrope and enters the body mainly *via* food. Another osteotrope, **lead-210** (**22.3 years**), is incorporated by inhalation though mostly by ingestion.

Thorium-232 is an alpha emitter with a half life of **14.1 thousand million years**. It possesses ten main alpha-, beta- and gamma-emitting radioactive descendants including **radon-220** (**55 s**). Thorium-232 enters the body mainly by inhalation. **Radium-228**, a direct descendant of thorium-232, is a beta-emitter with a half life of **5.75 years**. It enters the body mainly in food.

Radon, a gaseous radioactive descendant of uranium-238 and thorium-232, emanates from the soil and building materials, and along with its short-lived alpha-emitting descendants constitutes a source of internal exposure through inhalation. Radon is the most abundant source of natural radiation (about 40% of total radioactivity).

The human body contains nearly 4,500 Bq of potassium-40, 3,700 Bq of carbon-14 and 13 Bq of radium-226 essentially imported in food.

Natural radiation is supplemented by an anthropic component, resulting from the medical applications of ionising radiation and to a lesser extent from the nuclear industry. It accounts for about 14.5% of the total radioactivity worldwide, but much more in the developed countries. In the medical field (more than 1 mSv/year on average in France), irradiation by external sources predominates: radiodiagnosis (X-rays) and radiotherapy, long based on cæsium-137 and cobalt-60 sources, but now more and more often using linear accelerators. Irradiation by internal routes (curietherapy with iridium-192) has more specialised indications (cervical cancer, for example). The metabolic and physicochemical properties of some twenty radionuclides are put to use for **medical activities** and in **biological research**. The medical applications comprise radiodiagnostics (scintigraphy and radio-

(1) m for metastable. A nuclide is said metastable when a transition delay exists between the excited state of the atom and the stable one.

immunology), and treatment, including thyroid disorders using iodine-131, radioimmunotherapy in certain blood diseases (phosphorus-32) and the treatment of bone metastasis with strontium-89 or radiolabelled phosphonates alongside other uses of radiopharmaceuticals. Among the most widely used radionuclides are: **technetium-99m** (half life **6.02 hours**) and **thallium-201** (half life **3.04 days**) (scintigraphy), **iodine-131** (half life **8.04 days**) (treatment of hyperthyroidism), **iodine-125** (half life **60.14 days**) (radioimmunology), **cobalt-60** (half life **5.27 years**) (radiotherapy), and **iridium-192** (half life **73.82 days**) (curietherapy). The average contribution of radiological examinations to total radioactivity amounts to 14.2%.

The **early atmospheric nuclear weapon tests** scattered fallout over the whole of the earth's surface and caused the exposure of populations and the contamination of the food chain by a certain number of radionuclides, most of which, given their short radioactive half lives, have now vanished. There remain **cæsium-137** (**30 years**), **strontium-90** (**29.12 years**), some **krypton-85** (**10.4 years**) and **tritium** (**12.35 years**), and the isotopes of **plutonium** (half lives **87.7 years** to **24,100 years**). Currently, the doses corresponding to the fallout from these tests are essentially attributable to fission products (cæsium-137) and to carbon-14, rather than activation products and plutonium.

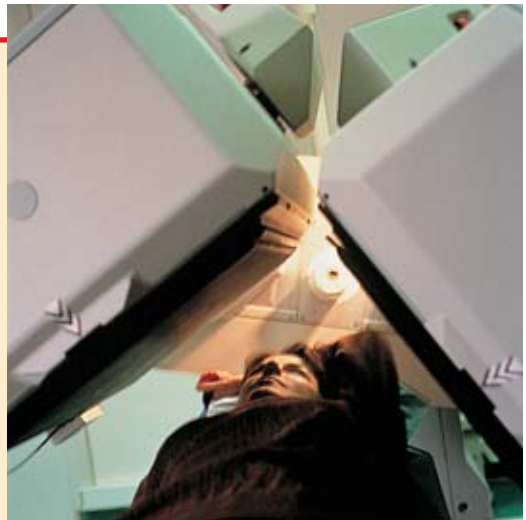
In the **Chernobyl accident** (Ukraine), which occurred in 1986, the total radioactivity dispersed into the atmosphere was of the order of 12 milliard milliard (10^{18}) becquerels over a period of 10 days. Three categories of radionuclides were disseminated. The first consisted of volatile fission products such as **iodine-131**, **iodine-133** (**20.8 hours**), **cæsium-134** (**2.06 years**), **cæsium-137**, **tellurium-132** (**3.26 days**). The second was composed of solid fission products and actinides released in much smaller amounts, in particular the strontium isotopes ^{89}**Sr** (half life **50.5 days**) and ^{90}**Sr**, the ruthenium isotopes ^{103}**Ru** (half life **39.3 days**) and ^{106}**Ru** (half life **368.2 days**), and **plutonium-239** (**24,100 years**). The third category was rare gases which although they represented most of the activity released, were rapidly diluted in the atmosphere. They were mainly **xenon-133** (**5.24 days**) and **krypton-85**.

The contributions of the early atmospheric nuclear weapon tests and the Chernobyl accident to the total radioactivity are roughly 0.2% (0.005 mSv) and 0.07% (0.002 mSv) respectively.

The whole of the **nuclear-powered electricity production** cycle represents only about 0.007% of total radioactivity. Almost all the radionuclides remain confined inside the nuclear reactors and the fuel cycle plants. In a nuclear reactor, the reactions that take place inside the fuel yield transuranics. **Uranium-238**, which is non-fissile, can capture neutrons to give in particular plutonium isotopes ^{239}**Pu**, ^{240}**Pu** (half life **6,560 years**) and ^{241}**Pu** (half life **14.4 years**), and **americium-241** (**432.7 years**). The main fission products generated by the fission of **uranium-235** (**704 million years**) and **plutonium-239** are **iodine-131**, **cæsium-134**, **cæsium-137**, **strontium-90** and **selenium-79** (**1.1 million years**).

The main radionuclides present in releases, which are performed in a very strict regulatory framework are, in liquid release, **tritium**, **cobalt-58** (**70.8 days**), **cobalt-60**, **iodine-131**, **cæsium-134**, **cæsium-137** and **silver-110m** (**249.9 days**). In gaseous releases **carbon-14** is the most abundant radionuclide, emitted most often as carbon dioxide. In all the reactors in the world, the total production of radiocarbon dioxide amounts to one tenth of the annual production formed naturally by cosmic radiation.

In addition, certain radionuclides related to the nuclear industry exhibit chemical toxicity (Box D, *Radiological and chemical toxicity*).



Laurence Médard/CEA

Classical scintigraphy performed at the Frédéric-Joliot Hospital Service (SHFJ). The gamma-ray camera is used for functional imaging of an organ after administration, usually by the intravenous route, of a radioactive drug (radiopharmaceutical) to the patient. The radionuclides used are specific to the organ being studied: for example, technetium-99m for the kidneys and bones, thallium-201 for the myocardium. The injected radiopharmaceutical emits gamma photons, which are captured by two planar detectors placed at 180° or 45° according to the examination.

B Human exposure routes

Human **exposure**, i.e., the effect on the body of a chemical, physical or radiological agent (irrespective of whether there is actual contact), can be external or internal. In the case of **ionising radiation**, exposure results in an energy input to all or part of the body. There can be direct **external irradiation** when the subject is in the path of radiation emitted by a radioactive source located outside the body. The person can be irradiated directly or after reflection off nearby surfaces.

The irradiation can be **acute** or **chronic**.

The term **contamination** is used to designate the deposition of matter (here **radioactive**) on structures, surfaces, objects or, as here, a living organism. Radiological contamination, attributable to the presence of **radionuclides**, can occur by the **external** route from the receptor medium (air, water) and vector media (soils, sediments, plant cover, materials) by contact with skin and hair (cutaneous contamination), or by the **internal** route when the radionuclides are **intaken**, by **inhalation** (gas, particles) from the atmosphere, by **ingestion**, mainly from foods and beverages (water, milk), or by penetration (injury, burns or diffusion through the skin). The term **intoxication** is used when the toxicity in question is essentially chemical.

In the case of **internal contamination**, the dose delivered to the body over time (called the **committed dose**) is calculated for 50 years in adults, and until age 70 years in children. The parameters taken into account for the calculation are: the nature and the intaken quantity of the radionuclide (RN), its chemical form, its **effective half life**[1] in the body (combination of **physical** and **biological half lives**), the type of **radiation**, the mode of exposure (inhalation, ingestion, injury, transcutaneous), the distribution in the body (deposition in target organs or even distribution), the radiosensitivity of the tissues and the age of the contaminated subject.

Lastly, the **radiotoxicity** is the toxicity due to the ionising radiation emitted by the inhaled or ingested radionuclide. The misleading variable called **potential radiotoxicity** is a *radiotoxic inventory* that is difficult to evaluate and made imprecise by many uncertainties.

(1) The effective half life (Te) is calculated from the physical half life (Tp) and the biological half life (Tb) by $1 / Te = 1 / Tp + 1 / Tb$.

F From rays to dose

Radioactivity is a process by which certain naturally-occurring or artificial **nuclides** (in particular those created by **fission**, the splitting of a heavy nucleus into two smaller ones) undergo spontaneous **decay**, with a release of energy, generally resulting in the formation of new nuclides. Termed **radionuclides** for this reason, they are unstable owing to the number of nucleons they contain (protons and neutrons) or their energy state. This decay process is accompanied by the emission of one or more types of **radiation**, ionising or non-ionising, and (or) particles. **Ionising radiation** is electromagnetic or corpuscular radiation that has sufficient energy to ionise certain atoms of the matter in its path by stripping electrons from them. This process can be *direct* (the case with alpha particles) or *indirect* (gamma rays and neutrons).

Alpha radiation, consisting of helium-4 nuclei (two protons and two neutrons), has low penetrating power and is stopped by a sheet of paper or the outermost layers of the skin. Its path in biological tissues is no longer than a few tens of micrometres. This radiation is therefore strongly ionising, i.e., it easily strips electrons from the atoms in the matter it travels through, because the particles shed all their energy over a short distance. For this reason, the hazard due to radionuclides that are **alpha emitters** is **internal exposure**.

Beta radiation, made up of electrons (beta minus radioactivity) or positrons (beta plus radioactivity), has moderate penetrating power. The particles emitted by **beta emitters** are stopped by a few metres of air, aluminium foil, or a few millimetres of biological tissue. They can therefore penetrate the outer layers of the skin.

Gamma radiation composed of high energy photons, which are weakly ionising but have high penetrating power (more than the **X-ray** photons used in radiodiagnosis), can travel through hundreds of meters of air. Thick shielding of concrete or lead is necessary to protect persons.

The interaction of **neutron radiation** is random, and so it is stopped only by a considerable thickness of concrete, water or paraffin wax. As it is electrically neutral, a neutron is stopped in air by the nuclei of light elements, the mass of which is close to that of the neutron.

• The quantity of energy delivered by radiation is the **dose**, which is evaluated in different ways, according to whether it takes into account the quantity of energy absorbed, its rate of delivery, or its biological effects.

• The **absorbed dose** is the quantity of energy absorbed at a point per unit mass of matter (inert or living), according to the definition of the International Commission on Radiation Units and Measurements (**ICRU**). It is expressed in **grays** (Gy): 1 gray is equal to an absorbed energy of 1 joule per kilogramme of matter. The *organ absorbed dose* is obtained by averaging the doses absorbed at different points according to the definition of the International Commission on Radiological Protection (**ICRP**).

• The **dose rate**, dose divided by time, measures the intensity of the irradiation (energy absorbed by the matter per unit mass and per unit time). The legal unit is the gray per second (Gy/s), but the gray per minute (Gy/min) is commonly used. Also, radiation has a higher **relative biological effectiveness (RBE)** if the effects produced by the same dose are greater or when the dose necessary to produce a given effect is lower.

• The **dose equivalent** is equal to the dose absorbed in a tissue or organ multiplied by a **weighting factor**, which differs according to the nature of the radiation energy, and which ranges from 1 to 20. Alpha radiation is considered to be 20 times more harmful than gamma radiation in terms of its biological efficiency in producing random (or **stochastic**) effects. The equivalent dose is expressed in sieverts (Sv).

• The **effective dose** is a quantity introduced to try to evaluate harm

Foulon/CEA

Technicians operating remote handling equipment on a line at the Atalante facility at CEA Marcoule. The shielding of the lines stops radiation. The operators wear personal dosimeters to monitor the efficacy of the protection.

in terms of whole-body stochastic effects. It is the sum of *equivalent doses* received by the different organs and tissues of an individual, weighted by a factor specific to each of them (weighting factors) according to its specific sensitivity. It makes it possible to sum doses from different sources, and both external and internal radiation. For internal exposure situations (**inhalation**, **ingestion**), the effective dose is calculated on the basis of the number of **becquerels** incorporated of a given radionuclide (**DPUI**, **dose per unit intake**). It is expressed in sieverts (Sv).

• The **committed dose**, as a result of internal exposure, is the cumulated dose received in fifty years (for workers and adults) or until age 70 (for those aged below 20) after the year of **incorporation** of the radionuclide, unless it has disappeared by physical shedding or biological elimination.

• The **collective dose** is the dose received by a population, defined as the product of the number of individuals (e.g., those working in a nuclear plant, where it is a useful parameter in the optimisation and application of the ALARA system) and the average equivalent or effective dose received by that population, or as the sum of the individual effective doses received. It is expressed in man-sieverts (man.Sv). It should be used only for groups that are relatively homogeneous as regards the nature of their exposure.

D Radiological and chemical toxicity

The chemical toxics linked to the nuclear industry include **uranium** (U), **cobalt** (Co), **boron** (B), used for its neutron-absorbing properties in the heat-exchange fluids of nuclear power plants, **beryllium** (Be), used to slow neutrons, and **cadmium** (Cd), used to capture them. Boron is essential for the growth of plants. Cadmium, like lead (Pb), produces toxic effects on the central nervous system. When the toxicity of an element can be both radiological and chemical, for example that of plutonium (Pu), uranium, neptunium, technetium or cobalt, it is necessary whenever possible to determine what toxic effects are radiological, what are chemical, and what can be either radiological or chemical (see *Limits of the comparison between radiological and chemical hazards*).

For **radioactive** elements with long physical **half lives**, the chemical toxicity is a much greater hazard than the radiological toxicity, as exemplified by rubidium (Rb) and natural uranium. Thus the chemical toxicity of uranium, which is more important than its radiological toxicity, has led the French regulators to set the **ingested** and **inhaled** mass limits for uranium in chemical compounds at 150 mg and 2.5 mg per day respectively, regardless of the **isotopic** composition of the element.

Certain metals or **metalloids** that are non-toxic at low concentrations can become toxic at high concentrations or in their radioactive form. This is the case for cobalt, which can be **genotoxic**, selenium (Se) (naturally incorporated in **proteins** or **RNA**), technetium (Tc) and iodine (I).

Cyrille Dupont/CEA

Two-dimensional gel electrophoresis image analysis carried out in the course of nuclear toxicology work at CEA Marcoule Centre in the Rhone Valley.