Jeremy Woodhouse/PhotoDisc

Sunflowers. The danger of human contamination by pollutants, irrespective of whether or not they are radioactive, derives partly from their entry into the food chain. The soil-plant transfer is an essential link in this entry.

Soil-plant transfer

Soil-plant transfer is an important link in the **contamination** of the human food chain by both **radioactive** and non-radioactive pollutants. This transfer is determined by many factors depending on the nature of the pollutant load, the soil composition and the physiology of plant species. In addition, these factors are sometimes inter-linked. The soil-plant transfer is often expressed by the **transfer factor**, defined as the ratio of concentrations in the plant or in a plant part, to those in the surrounding soil at equilibrium. There are a great many different transfer factors according to the pollutant, the soil, the plant species, the plant cover, the season(1) and the time elapsed since the pollution occurred. This simple parameter can be used to compare different elements or soil-plant systems, but provides no mechanistic insight or prediction of **bioavailability**.

The nature of the pollutant deposition, an important factor

The soil deposition of the pollutant, often uneven and superficial, with a low vertical penetration, can for a long time limit contact with roots and thereby root uptake. Conversely, marked accumulation of an element at the soil surface favours the contamination of the aerial parts through spattered soil. The assimilation of the element by *leaf absorption* is then possible – this happened after the Chernobyl disaster – but the consumption of the adherent soil is often a more significant route of entry into the food chain. The chemical and physical forms of the deposit are also decisive. Only elements in solution are mobile and interact instantly with soil components. The mobility of particulate deposits depends on the kinetics of dissolution, and so also on the properties of the soil, for example the **pH**, water content and **redox conditions**.

Soil composition, a key variable

To a large extent, the bioavailability of elements in the soil is a function of their mobility and therefore the ability of the soil to slow their movement in solution by a tortuous flow path, and above all to immobilise them by **adsorption**, reaction and **precipitation**. This immobilisation is often expressed by the *distribution factor* (*Kd*) defined as the ratio of solid phase concentration to solution concentration. The value of the distribution factor depends on the nature of the element and on the soil composition, in particular its pH, redox conditions, the nature of mineral and organic surfaces, and the concentration of homologous **cations** that compete for adsorption (Figure p. 10). For example, the distribution factor values measured for technetium range from near zero dm^3/kg, for pertechnetate (TcO_4^-) in soils in **oxidising** conditions, to several hundred dm^3/kg, in **reducing** conditions in soils rich in organic matter. Likewise, the distribution factor values for the radioactive **isotopes** of cæsium can fluctuate from less than 100 dm^3/kg for organic soils containing little clay to more than 10^5 dm^3/kg for clay-rich soils with a predominance of illites(2). Although a single distribution factor value is often measured for a given soil, it is important to note that a strong spatial and temporal variability of this factor is possible. In particular, the rhizosphere, a soil zone influenced by the action of roots and their associated microflora, differs from the rest of the soil. There are steep gradients of **redox potential**, pH and concentration of nutrients and pollutants. The result is a modification of exchange equilibria and sometimes a rapid alteration of soil minerals.

(1) The faster the vegetation grows, the more of a deposit is intaken.

(2) Illite: a potassium clay mineral with a foliated structure.

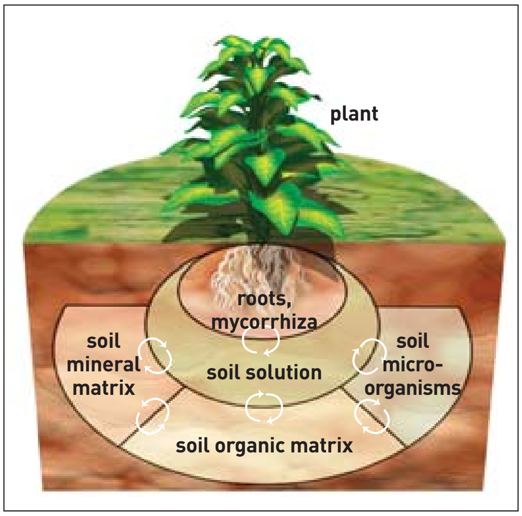


Figure.
The studies of transfer of radionuclides from the soil to plants take into account the different soil **compartments**.

Plant physiology, the third essential component

Lastly, the soil-plant transfer also depends on the plant species. The density, architecture and vertical root distribution determine the probability of a pollutant meeting a root. At the root-soil solution interface, pollutants and nutrients are adsorbed and absorbed by the roots. As with nutrition, the degree of mycorhization(3) influences the uptake of pollutants (Figure). However, the conclusions concerning the effect of mycorhizal infection on the contamination of host plants are often conflicting, because the pollutants can be stored in the hyphae(4). Root efflux(5) limits the accumulation on the roots and only a fraction of the element taken up is bound to the edible parts of the plant. All these processes depend on the plant species and its physiological state (age, nutrition, etc.). Whether the absorption is passive or active through specific carriers, the pollutants are taken up by the same mechanisms as nutrients – present in large or in trace amounts – that have similar properties. The competition among chemical species in biological membranes depends on the plant species and also on its physiological state and so on the number of active membrane carriers.

Radionuclides: limited assimilation

In general, **radionuclides** are present in very low chemical concentrations, and so are only weakly toxic, a notable exception being uranium. Thus there is less selection pressure favouring biological discrimination for radionuclides than for toxic chemical pollutants. The concentration of uranium in soils is insufficient for plants to develop systems to limit its assimilation. Recently molecular tools of plant physiology have allowed close study of the mechanisms that determine uptake and redistribution of pollutants in plants, thereby offering a better understanding of the origin of the variations in their bioavailability.

> **Siobhán Staunton**
National Agronomic Research Institute (Inra)
Montpellier (Hérault)

(3) Mycorhization: symbiotic association of a lower fungus with the roots of a plant.

(4) Hypha: a filament that is an elementary constituent of the mycelium (vegetative apparatus) of a fungus.

(5) Root efflux: excretory flow of materials from a plant through its roots to the soil.

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.

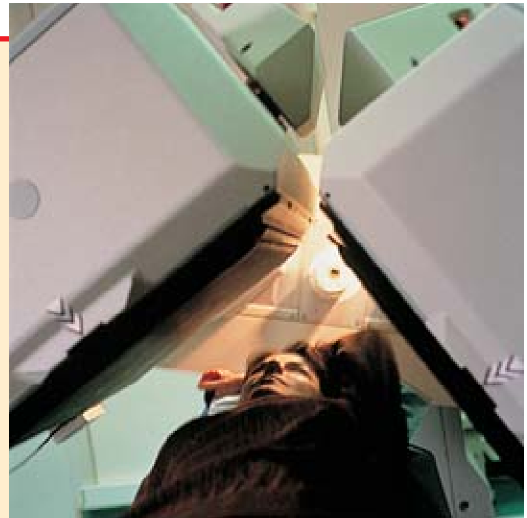
Thorium-232 is an alpha emitter with a half life of **14.1 thousand million years**. It possesses ten main alpha-, beta- and gamma-emitting radioactive descendants including **radon-220** (**55 s**). Thorium-232 enters the body mainly by inhalation. **Radium-228**, a direct descendant of thorium-232,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



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.

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*).

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

F (next)

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.