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Measurements of radioactivity in the environment for radiation protection purposes

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Summary. — Measurements of radioactivity in the environment, according to Article 35 of the Euratom Treaty, are routinely carried out by regulatory bodies and operators in charge. These activities are performed both locally and nationally, in order to assess effective doses to the population. In this context, therefore, it is necessary not only to determine the component due to external irradiation, but also to evaluate the deposit and accumulation of radioactivity in the environment that can reach the public via inhalation or ingestion. The contribution presents a brief description of the aspects regarding programs of monitoring and evaluation of the radioactivity in the environment, in particular the environmental matrices to be monitored, the analyses to be carried out, the related reporting levels as well as the features concerning the influence of the climate on the variations of dose rate values. Finally, a focus on some radiometric measurements performed over the last forty years in the environmental monitoring activities carried out in the ENEA Casaccia Research Center (Rome), in which two nuclear research facilities are currently operating, is presented.

1. – Introduction

Radioactivity is a natural component of the environment in which we live, contributing to the development of the earth's ecosystem since its origin, although it was only discovered at the end of the nineteenth century. Humans have always been exposed to natural radiation, including cosmic radiation, radiation emitted by radioisotopes contained in soils, rocks and building materials (*e.g.*, radioisotopes of the two primordial chains of ^{232}Th and ^{238}U and the primordial ^{40}K) and radiation present in food (mainly ^{40}K), as a consequence of the life cycle of plants and animals in the environment before

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being transformed into products for human consumption. Radioactivity can also have an anthropogenic origin (produced by human) including, for example, radiation generated by the production of nuclear energy, radiation used in hospital diagnostic and treatment activities (radiology, nuclear medicine and radiotherapy) and radiation present in the environment due to the fall-out of particles resulted by nuclear tests in the atmosphere (1960 - 1970) or nuclear accidents (Chernobyl in 1986 and Fukushima in 2011).

Recently, the international recommendations [1] published by the International Commission on Radiological Protection ICRP - the international body responsible for promoting the improvement of knowledge in the field of radiation protection - introduced the concept of *existing* exposure, for situations involving exposure to natural radiation sources([1]), and *planned* exposure, *i.e.*, linked to deliberate activities using radioactive sources or substances which are likely to increase the exposure of the population or the environment to ionizing radiation.

Exposure of the human body to ionizing radiation can take place in two ways: externally (*e.g.*, by stationing in a radiation field - the radiation source remains outside the human body) and internally (*e.g.*, by the introduction of one or more radioisotopes in the body mainly through inhalation and ingestion). In the international legislation [2], implemented in Italy with the Legislative Decree n. 101/2020 and further modifications, the *effective dose* quantity is used to estimate the increase of the risk of stochastic health effects (typically cancers) over the mean occurrence in a given population after external and/or internal exposure to ionizing radiation. The general criterion to consider activities involving the use of ionizing radiation (*practices* hereafter) exempted without further consideration contemplates, under all reasonably foreseeable circumstances, an effective dose equal or less than 10 μSv in a year, expected to be absorbed by any individual [3]. The value of 10 μSv in a year is considered a negligible effective dose, since it is associated to an increment of probability $p \approx 5 \cdot 10^{-7}$/year of risk of stochastic health effects [1]. For naturally-occurring radionuclides, the 10 μSv criterion is not generally applicable, since it is one or two orders of magnitude below the smallest value of exposure to natural background. In this case, the general criterion is to set the reference level of an effective dose equal to 1 mSv in a year [3] (increment of probability of risk $p \approx 5 \cdot 10^{-5}$/year of stochastic health effects [1]).

To evaluate the effective dose to the population it is necessary to quantify properly the radioactivity present in the environment. In this context, Article 35 of the Euratom Treaty [4] stipulates that *Each Member State shall establish the facilities necessary to carry out continuous monitoring of the level of radioactivity in the air, water and soil and to ensure compliance with the basic standards.* Indeed, measurements of radioactivity in the environment are routinely carried out by regulatory bodies and operators in charge, in order to protect members of the public from the risk of exposure to ionizing radiation. These activities are performed both locally, next to sites presenting nuclear or radiological risks, and nationally, through the so-called *environmental radioactivity monitoring networks*, geographically distributed across the national territory. These networks, consisting of measuring stations and sampling points, constitute an operational warning tool in case of an increase of the expected levels of natural radioactivity due to anthropogenic activities, such as an incident in one of the cross-border nuclear installations. They also

([1]) Existing exposure situations include also exposure to residual radioactive material that derives from past practices susceptible to no regulatory controls or from nuclear or radiological emergencies declared ended.

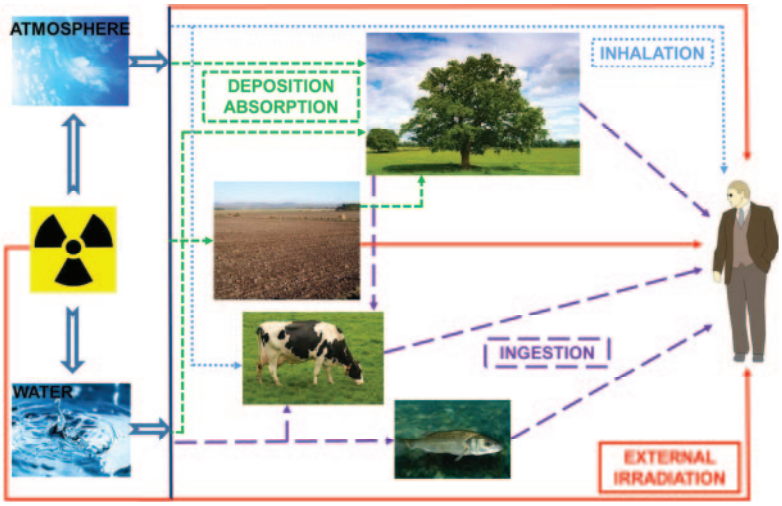

Fig. 1. – General representation of the environmental and food chains.

contribute to national databases, used for correct information to the institutions and the population, in order to maintain (or establish) public trust in the regulatory bodies and operators in charge for the common good.

Therefore, in the context of effective dose assessment, in addition to the determination of the component due to external irradiation and the identification of the different radioisotopes of interest to be considered, it is necessary to evaluate the deposit and accumulation of radioactivity in the environment that can reach the public via inhalation or ingestion. The routes through the different environmental compartments are called environmental and food chains. A simple schematic representation shown in fig. 1 provides a general picture of the phenomenon: the diffusion (controlled in authorized practices) of anthropogenic radioactivity in the environment can occur through dispersion in the atmosphere and/or in aquatic environments. Then the radioactivity is transferred to the soil, vegetation and crops through deposition and absorption phenomena. The transition to human being (in the broad sense to the animal species) can take place both by external exposure (*e.g.*, submersion in a radioactive plume or exposure due to radionuclides deposited on different surfaces as soil, buildings or vegetation) and by internal exposure, that can be direct, through inhalation of radioisotopes dispersed in the atmosphere, or indirect, by ingestion of food products of plant or animal origin. The evaluation of environmental and food chains, and therefore the evaluation of the extent of the transit to human being, requires a deep characterization of the environment, carried out through environmental monitoring with appropriate methods in relation to the characteristics of the territory of interest and to the habits of life and nutrition of the population.

In the following paragraphs, a brief description of the aspects regarding such programs of monitoring, in particular the environmental matrices and radionuclides to be monitored, the reporting levels as well as the influence of the climate on the variations of dose rate values, is presented. A focus is placed on some radiometric measurements performed over the last forty years on matrices sampled within the activities related to the environmental monitoring carried out in the ENEA Casaccia Research Center (Rome).

2. – Matrices and radionuclides

Dose assessment to the population is a multi-step process that must initially include:

- a complete characterization of the radiation sources to be used in the practice (*e.g.*, radionuclides, half-lives, type of radiation emitted),
- a characterization of the areas of the site selected for the practice, in terms of meteorological conditions (the historical map of the wind direction as well as the amount of annual rainfalls are of particular relevance) and current use of the territory (*e.g.*, the presence of livestock farming, crops and fishing),
- a characterization of the population surrounding the site, in terms of dietary habits and age distribution (*e.g.*, important for the identification of the *representative person*([2])).

The studies of this initial characterization should drive to a development of a dedicated sampling plan [5], involving all the aspects related to strategy and methods of sampling, transport and conservation of matrices, chemical-physical treatment and analysis of each radioisotope in each matrix, including the evaluation of the *best estimate* parameters, in order to provide an accurate environmental surveillance.

Environmental matrices to be sampled must include those for the control of the atmospheric diffusion term and those for the deposition in the environment. In particular some types of matrices to be monitored can be:

- air particulate, associated with human and animal respiration, to be sampled in densely populated areas with mostly no vehicular traffic, whose analysis gives the types and quantity of radioisotopes dispersed in atmosphere,
- fall-out, associated with the deposition of radioactive dust or fine particles from the atmosphere,
- surface water, underground water and marine water and sediments, associated with the spread of radioactivity in watercourses,
- soil, associated with the global quantity of radioactivity available for vegetation and crops,
- drinking water and foodstuffs (vegetables, meat, fish, milk and derivatives), associated with human and animal feeding.

The sampling plan typically includes monitoring stations, distributed in open field throughout the territory of interest, measuring the environmental gamma dose rate. The plan can also be integrated by the sampling of the so-called *sentinel organisms*, *e.g.*, mosses, mussels and brown algae in the sea or mushrooms on the land: normally they are not part of the diet of the population but they are important since are able to accumulate radionuclides at higher level with respect to other environmental matrices [5].

Concerning radionuclides, analysis should focus on radioisotopes used as radiation sources in the practice. Typically, the monitoring of the spatial-temporal trend of the radioactive concentrations in the environmental matrices is carried out in terms of:

([2]) An individual that receives a dose representative of the more highly exposed individuals in the population, excluding those individuals having extreme or rare habits [2].

- natural radionuclides, *e.g.*, ^{40}K and those originating from the thorium and uranium decay series (*e.g.*, ^{226}Ra, ^{222}Rn and ^{210}Pb), mainly associated with NORM and TENORM industries([3]),
- man-made radionuclides, *e.g.*, transuranic elements (*e.g.*, Am and Pu isotopes), ^{3}H, ^{14}C, Sr isotopes and gamma emitting radionuclides (*e.g.*, fission and activation products: ^{60}Co, ^{131}I and ^{137}Cs), mainly associated with nuclear activities (nuclear power plants and research reactors), radioactive waste repositories and hospital diagnostic and treatment activities.

3. – Dose-rate monitoring

As already mentioned, the sampling plan can typically include monitoring stations measuring continuously environmental gamma dose rates. Such measurements mostly play a key-role in environmental radioactivity monitoring since they provide, by setting dose rate alert levels, early warning in case of radiological or nuclear emergency.

Anthropogenic gamma-emitting radionuclides released in a potential emergency scenario can induce a suddenly increase of dose-rate value as soon as the detector used to monitoring the environment is submerged by the radioactive cloud during its passage. This peculiar footprint can be mimed also by weather phenomena, such as rain, snow and wind, which can influence the activity concentration of natural gamma-emitting radionuclides dispersed in air [6]. Unlike radionuclides in soil and building surrounding the detector, which can give an almost flat contribution to the dose-rate time series, radionuclides dispersed in air, such as ^{222}Rn (trough its short-lived gamma-emitting progeny ^{214}Pb, ^{214}Bi and ^{210}Tl), ^{7}Be and ^{40}K can contribute to sudden variations of dose-rate value especially during precipitations (rain-out and wash-out phenomena [7]). These dose rate variations typically develop in a hourly time-scale (at most few hours), so sampling times of 10 or 15 minutes (same of ones used for environmental monitoring) can be enough to appreciate the evolution of these phenomena. Unfortunately this timescale is comparable to the one characteristic of an anthropogenic cloud passage. The discrimination capability between natural phenomena and anthropogenic ones as causes of sudden dose-rate variations is then fundamental to detect any anthropogenic release.

4. – Reporting levels

In order to ensure compliance with the dose limits allowing for the protection of the public, the European Commission has established reporting levels on some environmental and food matrices (air, surface water, drinking water, milk and mixed diet), defined on the basis of their significance from an exposure point of view. They, indicated in table I [8, 9], are established both for specific radionuclides (^{3}H, ^{90}Sr and ^{137}Cs) and in terms of gross alfa/beta activity, mainly for screening purpose. The reporting levels are typically values evaluated with the exception criterion. Indeed, for air particulate and milk they are based on the 10 μSv criterion (for air particulate the representative person is an adult, breathing radioactive aerosols having a chemical composition with type S pulmonary absorption, whereas for milk the representative person is a newborn), instead the reporting levels for drinking water in terms of gross alfa/beta activity are

([3]) NORM: Naturally Occurring Radioactive Material and TENORM: Technologically Enhanced Naturally Occurring Radioactive Material.

TABLE I. – *Reporting levels for different sample types and different radionuclide categories* [8,9]. *Residual beta: the total measured beta activity minus* ^{40}K *activity.*

Sample types	Radionuclide categories	Reporting levels
Air	Gross beta (based on ^{90}Sr)	$5 \cdot 10^{-3}$ Bq/m^3
	^{137}Cs	$3 \cdot 10^{-2}$ Bq/m^3
Surface water	Residual beta (based on ^{90}Sr)	$6 \cdot 10^{-1}$ Bq/l
	^{137}Cs	$1 \cdot 10^{0}$ Bq/l
Drinking water	^{3}H	$1 \cdot 10^{2}$ Bq/l
	^{90}Sr	$6 \cdot 10^{-2}$ Bq/l
	^{137}Cs	$1 \cdot 10^{-1}$ Bq/l
	Gross alfa (screening)	$1 \cdot 10^{-1}$ Bq/l
	Gross beta (screening)	$1 \cdot 10^{0}$ Bq/l
Milk	^{90}Sr	$2 \cdot 10^{-1}$ Bq/l
	^{137}Cs	$5 \cdot 10^{-1}$ Bq/l

based on the effective dose equal to 0.1 mSv (the representative person is an adult). We emphasize that the reporting levels are not limits that cannot be exceeded, but they are just considered levels worthy of investigations if they are exceeded. No reporting level is defined for external ambient gamma dose rate, continuously measured.

5. – Local monitoring network for the ENEA Casaccia Research Center

An example of a local environmental radioactivity monitoring network in Italy is the one established and performed by ENEA for the Casaccia Research Center located in Rome (Italy), which covers an area with a radius of 5 km from the middle of the Center itself. Inside the Casaccia Research Center there are two operating nuclear research reactors (TRIGA and TAPIRO) and several facilities using ionizing radiation, including former fuel cycle research facilities and management of radioactive waste. The land around the Center is mainly used for agricultural crops and pasture. The main crops are represented by the production of fodder, wheat and cereals; the presence of orchards is minimal. The surface hydrography is constituted by the Arrone river, emissary of Bracciano Lake. The west part of the site is bordered by a little watercourse, which is the receiver of waste water of the Center and the radioactive liquid waste discharge; it flows into the Arrone river about 1 km from the southern end of the Center. The climatic characteristics of the Casaccia site reflect the climatic conditions of the regions of the middle Tyrrhenian Sea, with not exceptionally hot summers and mild winters. Winds are mainly northerly, with an average speed of about 1.5 m/s, 10 m from the ground.

The examined matrices are the following: air particulate, surface water, underground and drinking water, waste water, soil, sediment, grass and forage, vegetables, cereals and milk. The network also includes measurements of ambient dose equivalent H*(10) using TLD dosimeters, located throughout the site. The related measurement methods are: determinations of the concentration in gross alfa and beta activity, gamma spectrometry, alfa spectrometry for the detection of plutonium isotopes and measurements of ^{90}Sr. The sampling frequencies foreseen for the different matrices vary depending on the sampling points: daily (*e.g.*, atmospheric particulate and waste water), weekly/monthly (*e.g.*, sur-

face water and river sediment downstream of the site) and quarterly/half-yearly/annual (*e.g.*, milk, sediment upstream of the site, drinking water, soil and grass and forage). For vegetables and cereals, the number of samples varies according to local production.

5˙1. *Measurements on sampled matrices.* – Since 1985, about 2000 annual sampling of environmental and food matrices has been carried out. As an example, the trends of some radionuclide concentrations (^{90}Sr, ^{137}Cs and $^{239+240}$Pu) for the ENEA Casaccia site are reported in different matrices analyzed in the years 1985 - 2021. Specifically:

- for atmospheric particulate, the average annual concentrations of ^{137}Cs, compared with the reporting level indicated in table I [8], are shown in the left part of fig. 2. The maximum annual concentration value (0.02 Bq/m^3) was found in 1986 (the maximum daily concentration value equal to 2.2 Bq/m^3 was found in May, 1^{st}). All the remaining values are well below the established reporting level. In these thirty-seven years an annual concentration of ^{131}I above the minimum detectable concentration was found only in 1986 (0.1 Bq/m^3) and in 2011 ($1.4 \cdot 10^{-4}$ Bq/m^3).

- Regarding large leaf vegetables, typically chard, the plot in the right part of fig. 2 shows the annual concentrations of ^{137}Cs, whose maximum value (40 Bq/kg) was found in 1986. Analysis of ^{137}Cs provided in most cases concentrations below the related detection limit value; in the remaining cases, however, the concentrations remain comparable with the detection limit values of the measurement method.

- For milk, the average annual concentrations of ^{137}Cs and ^{90}Sr, compared with the reporting levels indicated in table I [8], are shown in fig. 3, left part and right part respectively. The maximum concentration values of ^{137}Cs (10 Bq/l) and ^{90}Sr (0.14 Bq/l) were obtained in 1986. All the ^{90}Sr values are below the established reporting level, instead the ^{137}Cs values remained above the reporting level for three consecutive years ($1986 \div 1988$). In the remaining cases the concentrations remain comparable with the detection limit values of the measurement method.

- Regarding soil, the average annual concentrations of ^{137}Cs and $^{239+240}$Pu are shown in fig. 4, left part and right part respectively. The concentration values of ^{137}Cs are in agreement with the values measured in the Italian territory (range $0.2 \div 2100$ Bq/kg). The maximum concentration value of ^{137}Cs (13 Bq/kg$_{dry}$) was obtained in 1987. In the absence of any European radiological events after Chernobyl, ^{137}Cs trend in soil respects the decay law with the corresponding half-life ≈ 30 years. Alpha spectrometry measurements on soil samples provided in most cases concentrations of $^{239+240}$Pu below the detection limit values; in the remaining cases, however, the concentrations remain comparable with the detection limit values of the measurement method.

In figs. 2, 3 and 4, for the years in which concentrations are lower than the related detection limits, the average values of the same detection limits are shown. For all annual mean concentrations shown here the associated uncertainty is equal to the standard deviation of the available data. The uncertainties associated with the individual measurements have not been taken into account here, because they vary according to the measurement technique (between 7% and 25%) and to the values themselves.

The extent and the trend of radiometric concentrations observed in recent years in the matrices sampled in the monitored Casaccia territory are compatible with both the events of Chernobyl in April 1986 and Fukushima in April 2011. On the other hand, the sporadic

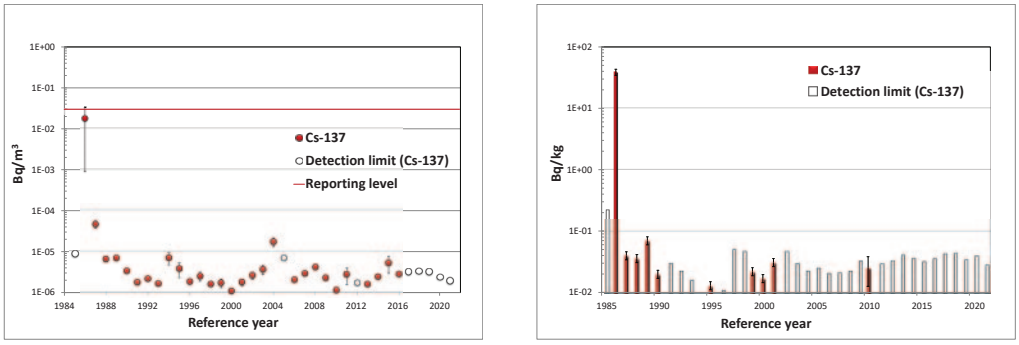

Fig. 2. – Left: average annual concentrations of ^{137}Cs in atmospheric particulate, sampled in ENEA Casaccia site. The reporting level for ^{137}Cs is also indicated [8]. Right: Average annual concentrations of ^{137}Cs in large leaf vegetables, sampled in ENEA Casaccia site.

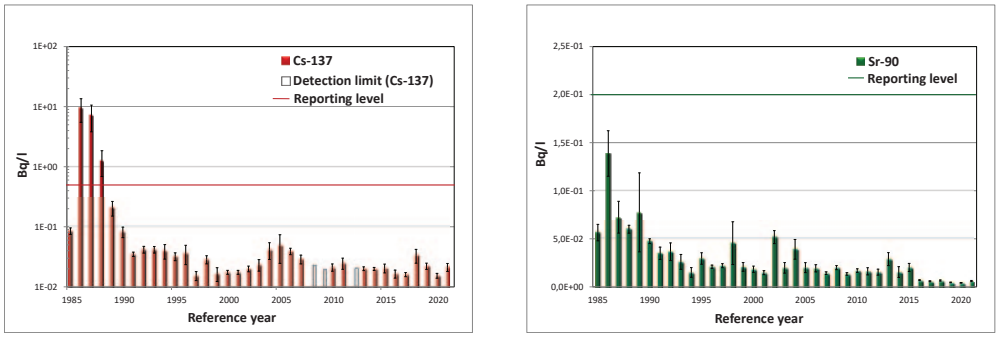

Fig. 3. – Left: average annual concentrations of ^{137}Cs in milk, sampled in ENEA Casaccia site. The reporting level for ^{137}Cs is also indicated [8]. Right: average annual concentrations of ^{90}Sr in milk, sampled in ENEA Casaccia site. The reporting level for ^{90}Sr is also indicated [8].

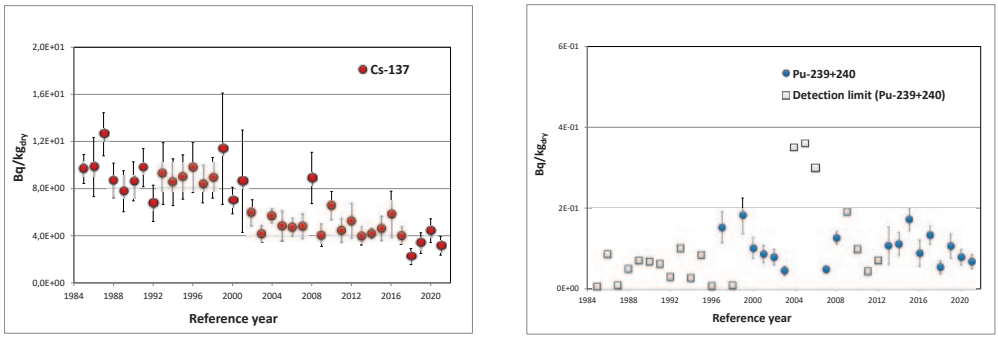

Fig. 4. – Average annual concentrations of ^{137}Cs (left) and $^{239+240}$Pu (right) in soil, sampled in ENEA Casaccia site.

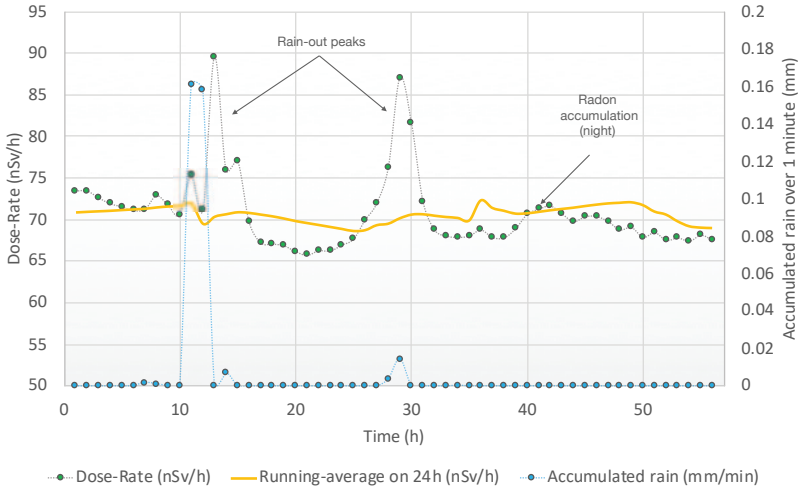

Fig. 5. – Typical dose-rate time series acquired with a sample time of 1 hour (green, colour on-line). In blue (colour on-line) is shown rain accumulated in a sample time of 1 minute and in yellow (colour on-line) is reported the running average on dose-rate values in a window of 24 hours.

presence of concentrations of $^{239+240}$Pu in environmental matrices, comparable with the values measured in the Italian territory, is attributable to the residual contamination of nuclear tests carried out in the atmosphere in the early '60s.

5˙2. *Dose rate measurements.* – Figure 5 shows a typical dose-rate time series with a sampling time of 1 hour. Peaks show a typical time duration of few hours with a shape compatible both with an anthropogenic cloud passage or with rain-out phenomena.

Data about the rain accumulated in 1 minute from a weather station, placed near the detector (shown in blue in figure), indicate that the peaks are generated by rain. When the rain starts to fall, it can drag to ground radioactive particulate in the cloud (rain-out) and the one dispersed in air below the cloud (wash-out), bringing it near to the detector and enhancing the dose-rate value measured by the detector. Peaks induced by rain-out shows a peculiar behaviour in case of heavy precipitations: the dose-rate values registered at the end of the rain event can be lower than the mean value registered before, because both the percolation of radioactive particles in soil and the radiation shielding of water permeating the soil. This peculiar behaviour is shown also during snow precipitations. In fig. 5 is also shown a typical structure due to radon night-accumulation phenomenon [10], detected by the monitor because the contribution of the short-lived gamma-progeny of ^{222}Rn. Radon exhalation from the soil is almost constant during the 24 hours, because it depends on ^{226}Ra content in soil. The atmospheric mixing layer, which acts as a cap for radon gas, varies its altitude from day (1-2 km) to night (few hundred meters). This variation of atmospheric volume available for radon diffusion causes a modification of radon concentration activity, which will be higher during the night (small volume) and lower during the day (larger volume). Therefore, to discriminate natural phenomena from anthropogenic ones in order to set-up dose rate alert levels with highest sensitivity, it is mandatory at least to use a weather station, to be coupled to dose rate detectors.

6. – Conclusions

In this work, aspects regarding programs of monitoring and evaluation of the radioactivity in the environment have been briefly discussed. Each Member State, according to Article 35 of the Euratom Treaty, must establish the facilities necessary to carry out continuous monitoring of the level of radioactivity in the environment, in order both to assess dose to the public and to identify events with possible impact on the environment and on the population promptly. In this context, therefore, it is necessary to monitor both the spatial-temporal trend of the anthropogenic radioisotope concentrations in the environmental matrices and the environmental dose rate. Concerning dose rate values, it is important to discriminate between natural fluctuations of environmental radiation (radon cycle, weather and precipitation) and anthropogenic phenomena.

A focus has been placed on some radiometric measurements performed on samples collected over the last thirty-seven years within the activities of environmental monitoring carried out in the ENEA Casaccia Research Center. Casaccia environmental data indicate that the activities carried out during the last years have not shown unauthorized radioactive releases into the environment, originating from nuclear plants present in the site. Consequently no increase in effective dose has been assessed to neighbouring population. Specifically, expect for values related to Chernobyl events, the concentrations obtained in atmospheric particulate, surface water, drinking water and milk have never exceeded the reporting levels established by European Commission (indicated in table I), while the concentration values obtained in foodstuffs, assuming that they are for Italian consumption, globally satisfy the 10 μSv criterion.

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