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SOME ASPECTS OF ENVIRONMENTAL RADIOACTIVITY RESEARCH

A. A. CIGNA

Laboratorio per lo Studio della Radioattività Ambientale, Comitato Nazionale per l'Energia Nucleare, C. S. N. – Casaccia, S. Maria di Galeria, Roma, Italia

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The differences between the situation before and after the »moratorium« of nuclear tests in 1962 are analyzed. Such differences led to different approaches to the problem of radioactive environmental contamination. The representativity of the measurements is discussed from the point of view of both the time and the site of sampling. Due to either the low level or the absence of radioactive contamination, the actual trend of the research concerns the study of the distribution patterns and the behaviour of stable elements in the biosphere.

Different methods of analysis are considered and a survey into the capacity of each of them is presented. The contribution of the ecological research in the field of radiation protection is discussed. Some indicators are well known by many researchers and proved to be valuable in the early detection of environmental contamination. The problem of the determination of the concentration factors is also considered. They play an important role in the establishment of the ecocycles of the radionuclides (fission and activation products) produced by nuclear power and research plants.

The reference levels of the radioactive contaminations in the items of the food-chain can be determined taking into account both the concentration processes involved and the specific activity.

The advantages of the different approaches are reported and discussed. Last but not least, the change in the radiation protection philosophy reported in the ICRP publication No. 7 is considered with the consequences in the development of the health physics techniques around nuclear plants.

The situation of the environmental radioactive contamination in the first sixties was peculiar, owing to the number and the power of the nuclear devices exploded in 1961 and 1962. A large number of measurements had to be performed in many countries in order to obtain sufficient information about the trend of a phenomenon which could be dangerous.

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Furthermore, such measurements were expected to be a valuable help in the establishment of theories on the propagation of radioactive contamination through the environment to man. From this point of view the work performed by Scott Russel's group of the Agricultural Research Council Radiobiological Laboratory and by Comar's group of the Cornell University, are particularly important. This work is a real guide, well known all over the world, to any researcher engaged in the field of environmental radioactivity. Among many correlation formulae concerning the radiocontamination in different items of the food chain, the most reliable is probably the one representing the relationship between dietary contamination and the deposition of strontium-90 (1):

$$C = p_r F_r + p_d F_d$$

where C is the ratio of strontium–90 to calcium in the food, F_r is the current rate of deposition, F_d is the cumulative deposit in the middle of the period under consideration and p_r and p_d are proportionality factors (»rate« and »soil« factors respectively). In the case of milk for a world-wide average $p_r = 0.8$ pCi Sr 90/gCa in milk per mCi Sr 90/km^2/years and $p_d = 0.3$ pCi Sr 90/g Ca in milk per mCi Sr 90/km^2 (cumulative deposit).

Such relationships give an estimate within the order of magnitude of the radioactive contamination of a foodstuff when the corresponding deposition of the contaminant is known. In the period following the »moratorium« the activity of the fallout due to the nuclear tests decreased to a very low level. In the meanwhile the new philosophy of health physics survey developed by many researchers emphasized the opportunity of the search of the »critical« nuclides and pathways.

The determination of the ecocycles of the radionuclides is the best approach to the solution of the problem. But on account of the reduction of fallout (the only tracer available in a geographical scale) the need for more accurate estimations became compulsory. The new trend is well represented by the ICPR Publication No. 7. According to the new principles presented in that publication, instead of performing a large number of measurements outside the boundaries of nuclear plants, it is more convenient to determine the »critical« nuclides and pathways. A measurement of the critical radionuclides in these pathways will be sufficient to survey the contamination in the environment because the values in the pathways will be an upper limit for the values in other places.

This peculiar behaviour of radioactive contaminations was clearly shown by experience and is also followed in large scale diffusion patterns. As an example, the concentration of strontium-90 in 15 sampling points in Italy during the period 1962–1967 can be considered (Fig. 1). The levels of Genoa and Varese are higher than the average of other sampling points. Therefore, from the point of view of pure protection from the Sr–90 fallout world wide contamination, a sampling network including Genoa and Varese only could be sufficient to give an upper limit of the average national contamination in Italy. Of course other reasons strongly support the opportunity to keep a nation-wide sampling network. The behaviour of the radionuclides may differ considerably according to their

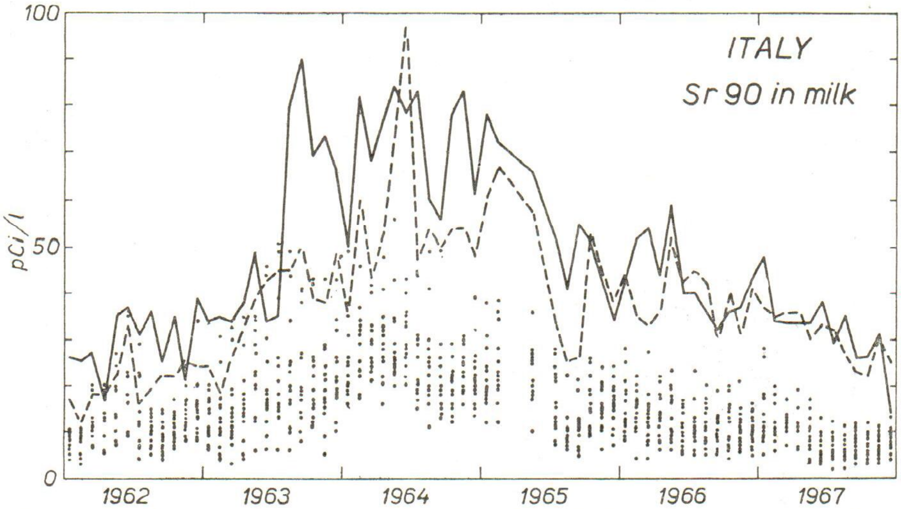

Fig. 1. *Strontium–90 concentration in Italian milk: full line refers to Genoa values, dashed line to Varese and dots to the other sampling points.*

chemical and physical properties. It is interesting to consider now the distribution of iodine–131 in milk. In Fig. 2 the concentrations in milk

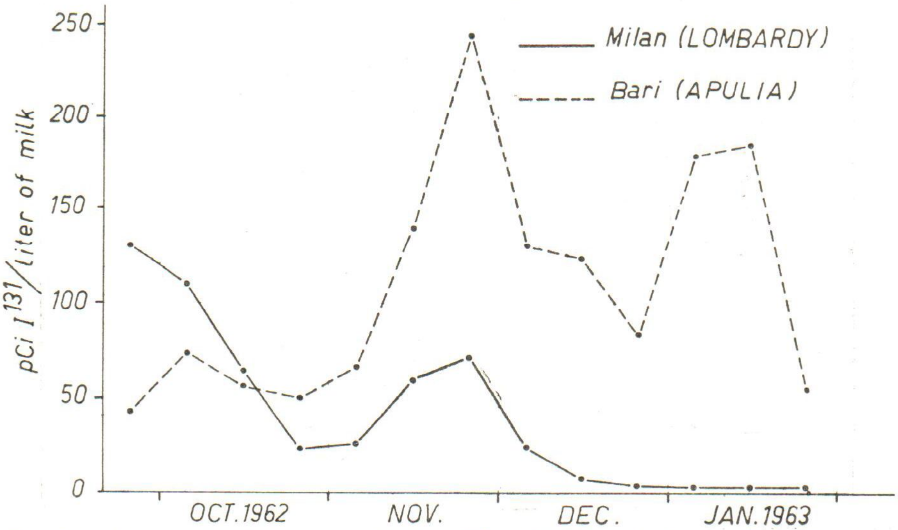

Fig. 2. *Iodine–131 in milk of two districts: Milan (Northern Italy) and Bari (Southern Italy).*

collected in Milan and Bari (2) are reported. The different patterns are due both to the oro-geographical features of the milk producing areas and to the feeding of animals with fresh fodder during winter months in the southern district (Bari). Local factors play an important role in the determination of the representativity of the sampling network with respect to the whole production of a foodstuff. In the case of milk the samples in Italy are collected in the distribution centers of 15 cities and are representative of the whole production of these centers. Such whole production has been called »measured« amount of milk. For each milk-shed a surrounding area has been defined, taking into account oro-

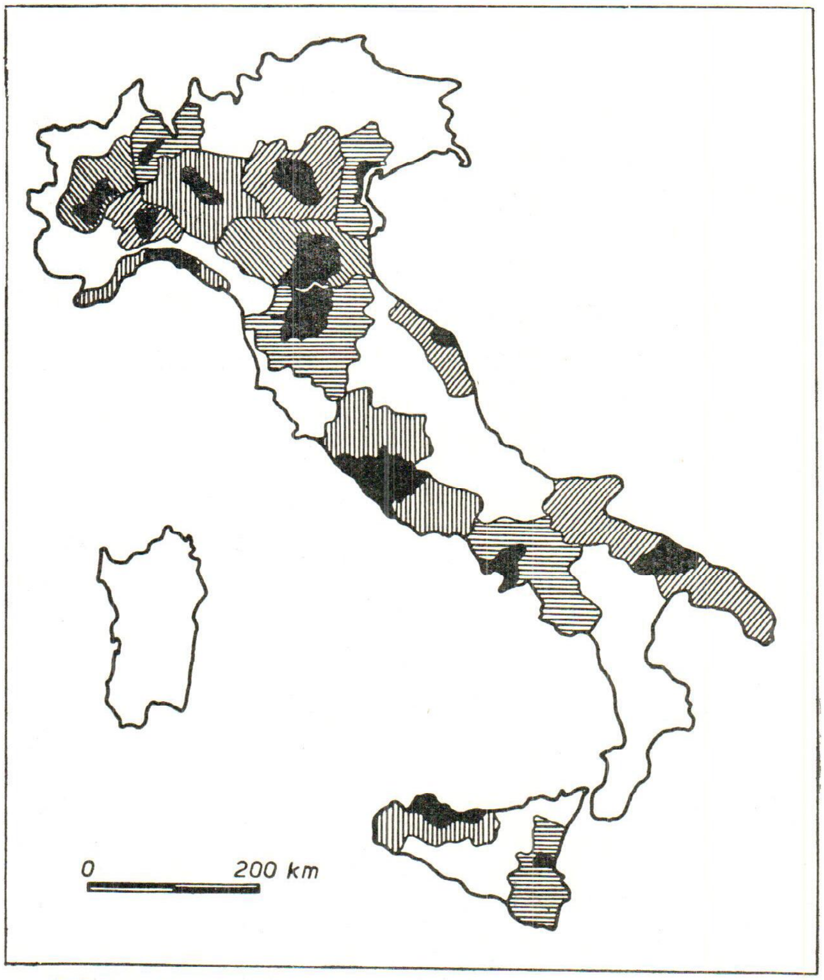

Fig. 3. *Milksheds (black) and surrounding areas (hatched) of the Italian milk sampling network.*

graphical, meteorological and agricultural features, so that the milk sample could be considered as representative for the defined area. The production of milk in such an area has been called »surveyed« amount. In Fig. 3 the milksheds and the surrounding areas are reported: the »measured« milk amounts to 14% and the »surveyed« milk to nearly 80% of the whole production in Italy (3). Of course these figures must be considered as approximate values averaged over a rather long period of time. A more detailed study on the representativity of the stations for the measurements of the atmospheric radioactivity in the German Democratic Republic (4) gave interesting results that about 80% of the activity variations of a measuring station in summer and about 60% in winter can be represented by those of the near-by stations, the mean distance between them being 130 km and on condition that fresh fission products are absent. The seasonal variation of the representativity is due to the meteorological differences, the air stability in winter contrasting the homogeneous distribution of the atmospheric contamination over a wide region. In Fig. 4 is presented a diagram correlating the percent-

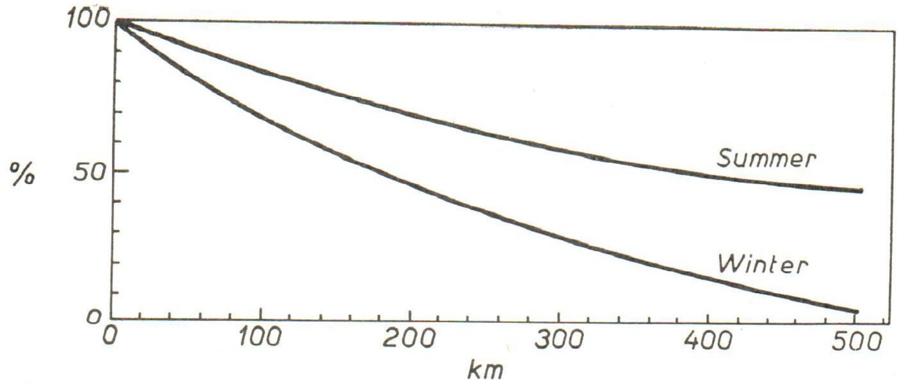

Fig. 4. *Seasonal correlation in German Democratic Republic between the percentage of activity variation of a given station represented by those of the near-by stations and the distance of the latter ones.*

age of the activity variation of a given station represented by those of the near-by stations with the distance of the latter ones. The diagram is valid for the German Democratic Republic, i. e. for a region without high mountains (altitude above sea level is always less than 1000 m for most of the region). Since January 1963 no important nuclear tests have been performed in the northern hemisphere. The Chinese and French bombs produced a small amount of radioactive fallout which did not change the general decrease of the activity observed in any country of the northern hemisphere. Therefore on account of the very low level reached recently by the radioactive contamination, the measurements of radionuclide concentrations in the environment became too difficult and affected by larger errors. For this reason the study of the distribution patterns and the behaviour of the stable elements in the biosphere became a more common practice. On account of the minor sensitivity of the analytical methods with respect to the radioactive measurements it became compulsory to use the most modern tools to measure the concentration of the stable elements in different samples of the biosphere. In Table 1 the sensitivities obtained with different methods for the most

Table 1

Sensitivity in μg/cm³

Element	Neutron activation (*)	Flame spectro-photometry	Atomic absorption	Absorption spectro-photometry	Oscillo-graphic polaro-graphy	X-ray fluorescence (**)
Na	0.00035	0.002	0.03	–	–	–
Mg	0.03	1	0.0005	–	–	–
K	0.004	0.01	0.1	–	–	2
Ca	0.19	0.03	0.1	–	–	2
Mn	0.00003	0.1	0.005	0.001	0.006	0.01
Fe	0.45	2	0.2	0.05	0.006	0.01
Co	0.001	10	0.013	0.025	0.006	0.006
Ni	0.0015	10	0.02	0.04	0.006	0.005
Cu	0.00035	0.1	0.005	0.03	0.006	0.04
Zn	0.002	2000	0.0005	0.016	0.006	0.02
Sr	0.03	0.1	0.05	–	–	0.03
Zr	0.015	–	9	0.13	–	0.03
Nd	0.5	20	250	50	–	0.05
Mo	0.005	30	0.5	0.1	0.01	0.5
Ru	0.005	10	0.3	0.2	–	–
Rh	—	1	0.3	0.2	–	–
Cd	0.0025	20	0.0004	0.01	–	0.7
Sb	0.0002	–	0.1	0.03	0.01	0.2
Cs	0.0015	1	0.15	–	–	0.1
Ba	0.0025	3	5	–	–	0.1
Pb	0.1	20	0.02	0.03	0.02	0.09
Bi	0.02	300	0.1	1	0.02	0.1

(*) 10^{13} neutrons/cm². sec.
(**) absolute amount in μg

common elements are listed (5, 6, 7, 8, 9). In general the neutron activation method gives the best results owing to both the high sensitivity and the small amount of material required. But it is necessary to use a nuclear reactor which is not yet a common equipment for any laboratory. On the other hand the X-ray fluorescence spectrometry seems to be very useful for fast and accurate measurements, the only difficulty being the high cost of the instrument. Other methods are cheaper but a little more time consuming. The best results can be obtained with a combination of the listed methods using the most suitable for each element to be investigated.

The search of the critical radionuclides and pathways is nowadays considered more useful than the establishment of a large number of sampling points randomly scattered in a region. The ecological research is the mean by which these critical radionuclides and pathways can be determined. In general the radioactive wastes from a nuclear plant are discharged into a river. Therefore the aquatic food-chain and the terrestrial one in the area where the river water is used for irrigation purposes, are studied. The work is commoly done by collecting at first data concerning the farm identification, characteristics, number of inhabitants, drinking and irrigation water supply, crops and cattle. A map of the region, where the different crops are reported, leads to the identification of the potentially critical crops on the basis of the concentration factors determined for each radionuclide and vegetal species. In other words, the ecocycles of the radionuclides are determined. At present these ecocycles are known only rather roughly because very often the concentration factors are estimated for lack of direct evaluations. A detailed knowledge of the concentration factors for the most interesting species in different environmental conditions is an important field of research for the ecology applied to environmental radioactivity. The determination of »real« concentration factors is sometimes difficult as the equilibrium conditions are required. In many cases the concentration of a radionuclide in the biosphere is subject to irregular variations (due

Table 2

Dry land environment

Date	Place	Material	Element	FC (*)	Reference
1962	Austria	Peltigera canina	Sr	10	(***)
1962	Austria	Peltigera canina	Cs	7	(***)
1960–61	Hungary	Helix pomatia	Sr	30	10
1961–63	Italy	Thyroids of cattle	I	10 ÷ 500(**)	(***)

(*) with respect to the soil
(**) with respect to the milk
(***) unpublished data

to the rate of deposition, soil absorption, dilution, etc.) and the metabolic processes introduce a phase lag of the corresponding variation in the organism under examination. Therefore a comparison of the concentration of the radionuclides in the organism and in its environment may also give a value of the »observed« concentration factor which is quite different from the value obtained in equilibrium conditions. To avoid this inconvenience either the number of determinations should be increased (in order to average the varying conditions) or the determination must be performed under well-known steady state conditions (13).

The determination of the concentration factors is useful for another purpose too, i. e. for the search of the so called biological or ecological indicators. These are organisms (and somtimes inanimate materials, as sediment, tea-bags, etc.) characterized by a high value of the concentration factor. Some indicators for each ambient can be remembered. In Tables 2, 3 and 4 some examples are reported (10 to 18). The indicators sometimes proved to be the only tool for detecting the presence of a radionuclide in the environment. In 1960 the contamination due to manganese – 54 in Maggiore Lake (Northern Italy) water was discovered for the first time by molluscs measurements when the radioactivity of the water was well below the detectable minimum (11).

Table 3

Fresh water environment

Date	Place	Material	Element	FC	Reference
1960–61	Italy	Unio pictorum	Mn	$10^5 \div 4 \times 10^5$	11
1960–61	URSS	Mougeotia, sp	Co	2×10^5	12
1963–65	Italy	Perca fluviatilis	Cs	$460 \div 14700$	13
1963–65	Italy	Scardinius erithrophtalmus	Sr	$84 \div 1040$	13
1961	U. S. A.	Unionidae	Sr	$4800 \div 6500$	14

Table 4

Marine environment

Date	Place	Material	Element	FC	Reference
1961–63	Italy	Plankton (20% of Achantaria)	Sr	$180 \div 390$	15
1960–61	Great Britain	Cartilage of Raia clavata	Sr	16	16
1962	U. S. A.	Enteromorpha, sp	Mn	1000	17
1958	U. S. A.	Ommnastrephes	Fe	10^4	18
1958	U. S. A.	Ommnastrephes	Cd	2×10^5	18
1958	U. S. A.	Calanus	I	100	18

Sometimes the determination of the Derived Working Limits (DWL) in the different media of the food-chain is rather cumbersome because of the many processes taking place during each transfer of contamination from one item to another. Two approaches can be followed (19). They have different advantages and therefore the proper one can be chosen according to the specific situation. The first approach (20) takes into consideration the intake of radionuclides to man by ingestion of foods. Therefore it is possible to establish a value for the concentration of radionuclides in the medium under examination below which the concentration in the succesive steps of the food-chain will not lead to greater than allowable intake by man. The value calculation is based upon the concentration factors in the steps of the food chain taking into account, of course, the Maximum Permissible Concentration (MPC) given by the ICRP and man's daily intake of a certain food.

The second approach (21), which is also known as »the specific activity approach« is based upon the principle that the allowable amount of a radionuclide in man cannot be exceeded by ingestion of any quantity of food if the specific activity (i. e. the ratio between the concentration of a radionuclide and the concentration of the stable element) of the medium in which the food was grown was no greater than the allowable specific activity for man. Two underlying assumptions to this approach are that there is no isotope separation (between radioactive and stable isotopes) in the metabolic processes and that the physical and chemical forms of the stable and radioactive isotope in the medium are the same.

Both methods are valuable: the former requires a knowledge of the ecocycles of the radionuclides (i. e. the FC involved in each step of the food chain) but it is independent of the abundance of the element under consideration in the biosphere. On the other hand, the latter requires no information on the processes taking place in each step but gives reliable results only when the stable elements are present in the biosphere in non-negligible amounts. The concept of the specific activity can be widened to consider the dilution of the radionuclide due to the whole amount of the isometabolic elements. In the case of strontium–90, e. g., the activity should be referred to the correspondent amount of stable strontium and calcium. In many cases the contribution of stable strontium can be neglected in comparison with the contribution of calcium and therefore the so called »strontium units« (pCi Sr90/g of Ca) can be used. On the same basis the »caesium units« (pCi Cs 137/g of K) were introduced some years ago, but there are many objections to a common use on account of the discrepancies between caesium and potassium metabolism. Another consequence of the specific activity concept is the possibility of a further isotopic dilution of the waste disposal from nuclear plants which can be achieved by adding suitable amounts of the stable elements whose radionuclides are included into the waste. The chemical toxicity of these elements is the limiting factor to the addition of the carriers but sometimes a considerable dilution factor can be obtained with low concentrations of stable carriers. Sometimes when the chemical toxicity is rather low, the factor limiting the amount of radioactive waste can be given by direct irradiation. The change in health physics philosophy as reported in ICRP publication No. 7 must not be overlooked or considered from the wrong point of view. The possibility to reduce the number of sampling points around a nuclear plant is established with some assumptions which cannot be forgotten. An example of the evolution of the sampling network around the CNEN Center of Nuclear Studies of Casaccia (Roma) will emphasize this concept. When the network reached its maximum size in 1964 a fairly large number of sampling points was spread all around the Center area (Fig. 5) to cover many items of the food chain

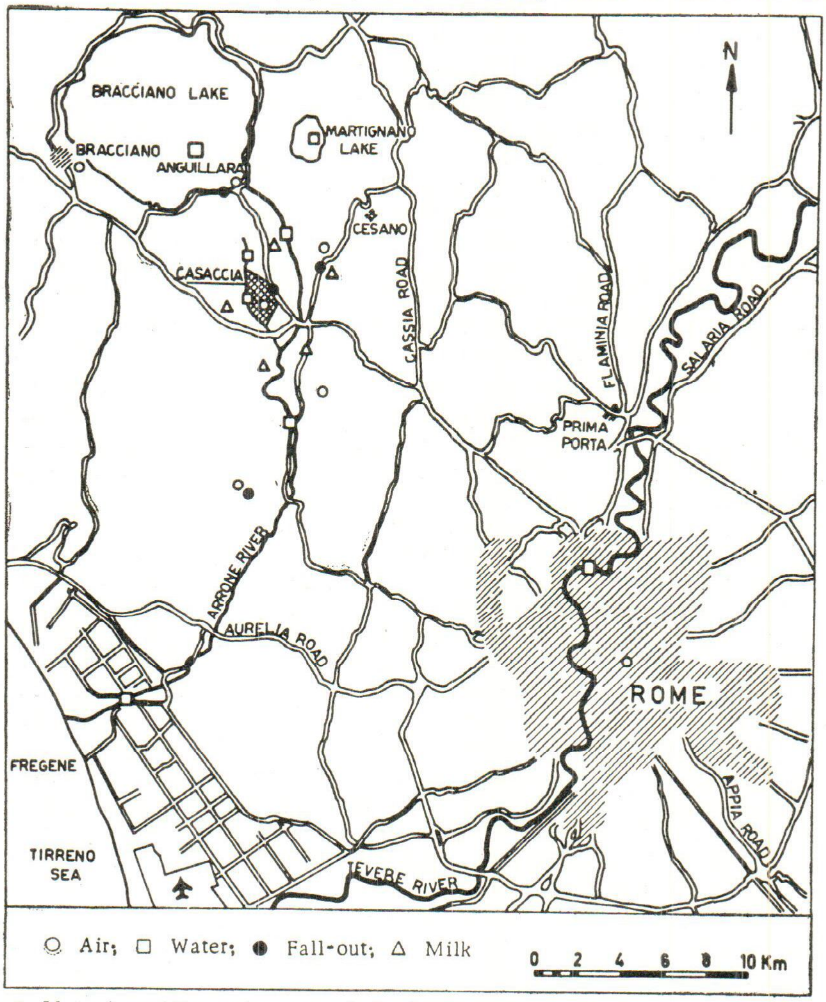

Fig. 5. *Map of sampling points around the Casaccia Nuclear Studies Center near Rome.*

also (22). The recent ecological investigations (23) have led to the identification of two critical areas when the critical pathway is the ingestion of contaminated foods (Fig. 6). At present the sampling points are not drastically reduced to cover the two critical areas only, because of the necessity to survey the consequences of an unwanted release of conta-

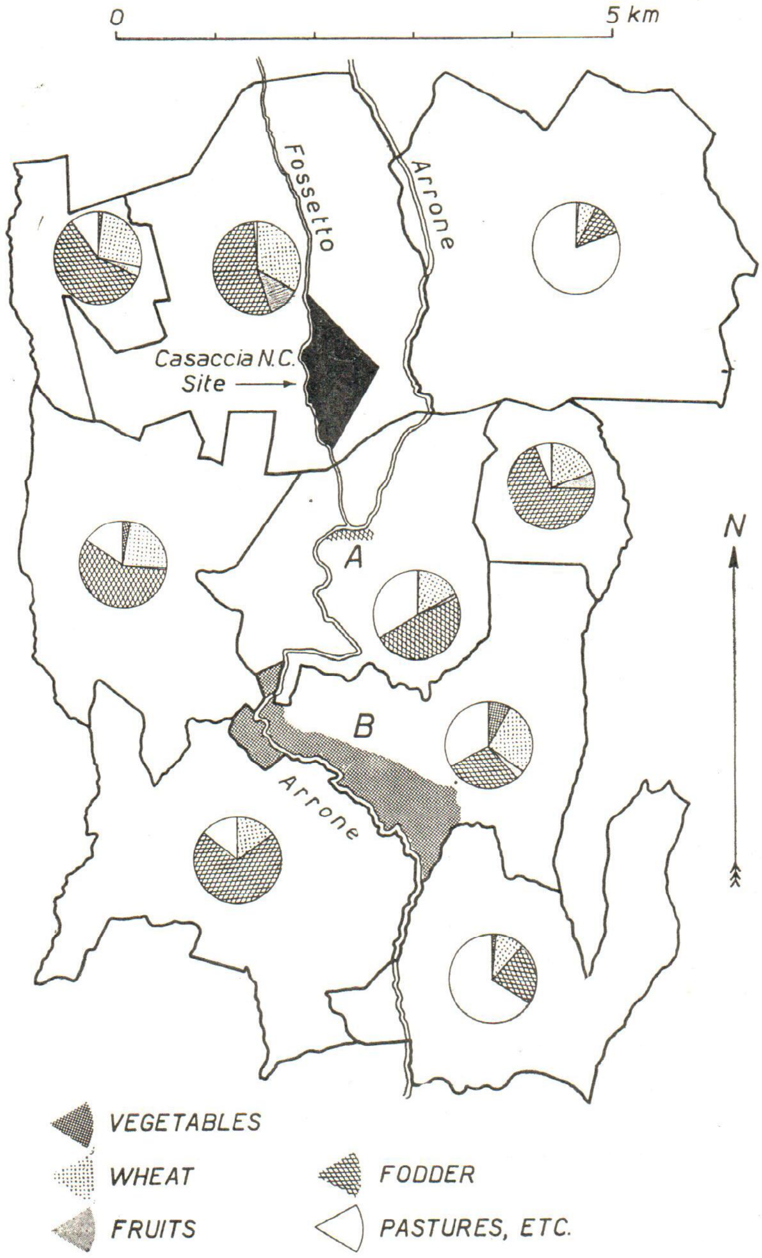

Fig. 6. *Map of the critical crops (A: fodder field; B: vegetable field). The percentage of the different cultivations in each group of farms are also reported.*

mination which may not follow the pattern considered in the ecological investigation, but a suitable decrease of the sampling points number was allowed.

Indeed, this research was based upon the normal disposal of low level radioactive wastes into a nearby small stream whose water is used for irrigation purposes. On the other hand the identification of the critical areas was determined by the distribution of crops in the surroundings of the Center. Any change in the type of cultivation may induce a change in the determination of the critical areas. In other words an ecological survey cannot be considered »dead« when all the data are collected but it is always »alive« and must be kept up to date with the variations occurring in the region under consideration. One of the founders of radiation protection, dr. *K. Z. Morgan* wrote: »Many factors, some imponderable, contribute to the estimation of a maximum permissible dose« (24). It is the particular feeling of these imponderable factors which turns the profession of the health physicist into an art.

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Sadržaj

NEKI ASPEKTI ISTRAŽIVANJA RADIOAKTIVNOSTI OKOLINE

U radu je analizirana razlika u općoj situaciji prije i poslije moratorija nuklearnih pokusa 1962. godine. Zbog veće radioaktivne kontaminacije hrane prije moratorija, bilo je moguće postaviti odnos između kontaminacije hranom i depozicije Sr-90 u ljudskom organizmu. Nakon moratorija aktivnost padavina je svedena na minimum. Uslijed toga došlo je do drugačijeg pristupa problematici kontaminacije životne sredine. Sada se više polaže pažnja na mjesta odakle se uzima uzorak i na reprezentativnost mjerenja pojedinog uzorka s gledišta utroška vremena na pojedinu analizu.

Osim toga, bilo zbog niske aktivnosti biosfere bilo zbog odsutnosti radioaktivne kontaminacije u njoj, sadašnja su istraživanja usmjerena na ispitivanje ponašanja stabilnih elemenata u biosferi i na način njihove distribucije u ekološkom ciklusu. Pri tome je dana ocjena primijenjenih kemijskih i radiokemijskih metoda za njihovo određivanje, pregled metoda i granice osjetljivosti.

Prikazan je udio ekoloških istraživanja na čitavom području radiološke zaštite. Traže se indikatori za ranu detekciju kontaminacije okoline. Takav indikator može biti jedan od »kritičnih«, npr. Sr-90, kojega je praćenje kroz ekološki ciklus dovoljno da se dobije faktor kontaminacije. Pritom je izbjegnuta obrada velikog broja uzoraka, pa se, dakle, smanjuje kontrolna mreža i lakše se odredi faktor kontaminacije.

Faktor kontaminacije je važan pri utvrđivanju ekocikla radionuklida (fisionih i aktivacionih produkata), proizvoda nuklearnih centrala ili istraživačkih postrojenja. Za sada su ti ekocikli samo grubo poznati. Referentni nivoi radioaktivne kontaminacije, potrebni za određivanje koncentracionog faktora u prehrambenom ciklusu,

određeni su, naime, na temelju vrijednosti koje variraju, a ne u ravnotežnim uvjetima – okolina – čovjek – klima. Zbog pomanjkanja direktne a točne evaluacije, koncentracioni faktori procijenjeni su više empirijski.

Rad završava evaluacijom različitih pristupa problematici radioaktivne kontaminacije životne sredine. Posebna se važnost pridaje izmjeni načina ekoloških ispitivanja nakon objavljivanja publikacije ICPR br. 7 i kroz to nastalih posljedica. Mijenja se tehnika radiološke zaštite oko nuklearnih postrojenja.

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Laboratorij za proučavanje radioaktivnosti okoline, Nacionalni komitet za nuklearnu energiju, C. S. N. – Casaccia, S. Maria di Galeria, Rim, Italija