

Goal Oriented Sampling Strategies for Radionuclide Monitoring

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Abstract

This work carried out in 2014 in the frame of Institute of Environmental Radioactivity at Fukushima University. Based on the experience gained during the performing countrywide monitoring of radionuclides in Hungary a sampling schedule was suggested for assessing the dose for the public in Japan. Many specialities are existing due to the size, climate, traditions which has to be taken into account: for establishing the long term series of the data the suggestion is to monitor in each prefecture: rice leafy vegetable, like spinach, root vegetable, seaweed, soy bean. For having well coverage in terms of spatial distribution, same sample across the whole country has to be collected: moss as integral indicator found everywhere in the country, fresh leaves of bamboo could be treated as differential indicator, bamboo spot forests in different sizes are found in the country. Bamboo leaves are proved to be a proper reference material in dried state. Above the samples the installation a large volume aerosol filter with about 250m³/h airflow and big filter surface (larger than 0.2m²) would be desirable in each prefecture. Several pilot studies were conducted and written as case studies and lessons learned were summarised as a suggestion.

Keywords

monitoring, sampling, radionuclides, Fukushima

1.0 Introduction

There is a need for concise general guidance on optimum environmental sampling for radionuclide studies, including drawing attention to possible misinterpretation of the results of a survey. Inadequate monitoring strategy can greatly affect the effectiveness and even the quality of the emergency response, the evaluation the situation crossover the country and the scientific interpretation. There is no doubt on variation of environmental media, it is an ever present issue independently on the way of sampling, e.g. soil taken side by side or different parts of the vegetation or sample is taken from same type of animals living in the same type of environment. Therefore the variability is caused by natural variations in the radionuclide transport and uptake governed by environmental processes has to be controlled using some statistical approach. Individual sampling units are always unique; therefore taking of a sufficient number of units can provide a proximately true estimation or reflection of the examined environment. The representativeness of the environmental sampling is difficult to demonstrate; just well-defined sampling protocols could give information about. Sampling strategy has to contain at least a plan of place and timing of the sampling, what kind of samples and how many sampling unit in what amounts has to be collected. The purpose of the survey defines the sampling strategy whether that be for compliance purposes, normal regulatory or emergency management. Finding out the best sampling strategy needs thorough knowledge of the environment where sampling has to be carried out. The applied sampling

strategy should be flexible enough to take care about the variability of the given environment and to handle the unexpected situations.

For elaborating a proper strategy the objectives has to be defined, the properties of the surveyed environment have to be known well. For the representativeness of sampling is important to identify the target population. The appropriate sampling protocol has to be elaborated and applied; when the radioanalytical results are available and summarised the feedback for improving the whole strategy is necessary. Under a probabilistic sampling strategy it is assumed that the population can be specified that each member of the population has probability of being selected.

After a short guidance the lessons learned are shown through several case studies for providing some solutions or giving examples even for not really perfect programs.

2.0 Optimisation of the Monitoring

Rather hard to give proper guidance on all kind of special cases of monitoring programs, sampling strategies and conducting of any particular kind of study, but it is possible to summarize and give the guidance on what has to be take into account and what are the main influencing effects for defining their importance in that particulate case. Above the properly defined objective of monitoring the population characteristics in statistical meaning also has to be taken into account, simple random, stratified random and systematic sampling is applied most commonly. The last is applied for monitoring over time mainly, when samples are taken in the regular intervals. The multi-phase strategy also should be considered.

Never forget about the components and causes of the variability of the results of a survey; the variability includes:

- natural variability of the samples is the result of place-to-place, time-to-time and individual sample-to individual sample,
- variation in the designed monitoring, in sampling strategy, in sampling protocol,
- variation of treatment of the samples and applied radioanalytical techniques.

Many years of experience showed already that usually the natural variability dominates over the variability of the laboratory analysis of the samples in a steady period without unusual contamination. Above the contamination processes the radioecological processes are also inherently complex in space, time and element of environment and target radionuclides causing further spatial heterogeneity. It is not ordinarily possible to ascribe observed variability to any particular sources. From the literature review comes that the variance partitioning was seldom quantitatively revealed, although in nearly all cases the dominant source of uncertainty appeared to be associated with inherent variations resulting from spatial or individual organism or sample variability. It is typical that the analysis replication variation is just 1% of the spatial sampling variance and the sampling variance is in the order of 20% as it is analysed in ICRU report 75 [1]. The data reviewed also show a tendency for vegetation to be less variable than in most cases. Regarding the variance of the terrestrial animals there is a difference even between the different organs, and in a spatially heterogeneous environment individuals show considerable variability by using only certain feeding and area. The variability strongly depends on the radionuclide too, e.g. plutonium isotopes, americium isotopes are heterogeneously distributed because of their chemical properties (low solubility, moreover sometimes the contamination is just on the surface and not incorporated into the body). Animals of aquatic ecosystems illustrate usually well that the variability is affected by the behavioural pattern too.

Scott et al. (2008) [2] suggest five-step method for defining the proper strategy. ICRU report 75 [1] also deals with the sampling for radionuclides in the environment. Both of the

documents provide nice details about the topic. Design of sampling strategy requires problem-specific environmental knowledge, statistical knowledge practical knowledge concerning implementation of sampling and knowledge about the radioanalytical techniques. Summarising and complying the thoughts from monitoring point of view the decision has to be made thinking over the circumstances and facts in the following order:

1. Objective of the monitoring or sampling (multiple objectives are not rarely defined especially in case of a monitoring program):
 - 1.1. defining background levels,
 - 1.2. describing the magnitudes of variability,
 - 1.3. observing temporal trends,
 - 1.4. describing spatial patterns,
 - 1.5. mapping,
 - 1.6. detecting contamination above background level,
 - 1.7. assessing compliance with regulations,
 - 1.8. assessing environmental impact of a facility,
 - 1.9. assessing environmental impact of an event,
 - 1.10. determining the characteristics of a population.
2. Identification of target or targets
 - 2.1. the radionuclide or nuclides to be measured,
 - 2.2. part or parts of the environment to be investigated,
 - 2.3. the target population,
 - 2.4. complex sampling – e.g. any kind of food-chain.
3. Collect information about the target population as much as possible
 - 3.1. environmental properties,
 - 3.2. meteorological conditions,
 - 3.3. spatial information, like land use map, map about soil properties,
 - 3.4. spatial density of the population,
 - 3.5. environmental conditions having special effect on the population has to be studied more thoroughly,
 - 3.6. erosion map, density of any kind of disasters (e.g. earthquake map).
4. Choosing an effective, fit-for-purpose sampling design
 - 4.1. systematic sampling,
 - 4.2. stratified random sampling – complex, prior knowledge,
 - 4.3. random sampling, two-stage sampling,
 - 4.4. judgemental sampling,
 - 4.5. cluster sampling,
 - 4.6. multiple sampling or sample pairs from the same location,
 - 4.7. double sampling.
5. Careful consideration about the data has to be noted during the sampling
 - 5.1. code of the individual sample,
 - 5.2. location,
 - 5.3. date,
 - 5.4. dose rate,
 - 5.5. topography, landscape,
 - 5.6. any kind of physical, chemical, biological properties,
 - 5.7. any kind of influences might have effect on the distribution of the radioanalytical results (eg. weather),
 - 5.8. remarks, photos, videos – they might provide essential information or just additional information during the evaluation phase.

A prior environmental study is strongly recommended for collecting as much as information possible for example with a simply round trip on the field. This knowledge will prevent the mis-pointing the sample area and sampling points, which might result a really cost effective solution at the end of the survey. Sometimes even the aerial survey might be economical if the area is large enough. Careful consideration of the circumstances should lead to representative sampling, however representativeness of environmental sampling is hard to give, usually it has to be justified by the sampling procedure elaborated. Estimating the variance of the population might be with pilot study, with previous sampling of the same or similar population, with relying on the scientific literature, with scientific judgement which in fact might be subjective. If there is almost no knowledge for the desired program worth to conduct a pilot study for getting information and the order of magnitude of variation.

Never forget about the comparability and consistency of sampling and measurement protocols in case of a bigger survey when many institutions and laboratories are involved, they have to be well harmonised.

An example is shown on the 1. Figure about the influencing factors in the planning a survey – any kind of diagram is worth to use for the visualisation and for the better understanding of the needs and presenting it to the decision makers.

Basics for executing the sampling program:

In normal situation the sampling team is only two colleges, in case of emergency the minimum number of team members is three:

Person A executing the sampling - he/she is the team member “getting dirty”,

Person B gives the prepared tools to Person A and take care about avoiding the cross-contamination between samples,

Person C takes care about the administration, such as writing the sampling report, marking and arranging the samples which are already in clean plastic bag.

Two apparatus are always necessary for sampling:

1. Dose rate meter capable to measure low doses. There is some equipment available which has a main unit for measuring high dose-rates and connected to an additional probe (mainly scintillation probe) is able to detect doses from 10nSv/h.
2. GPS capable to mark the sampling point and give the longitude, latitude and elevation data. some GPS is able to determine the area also which might be useful, especially in an unknown field.

The additional batteries for both apparatus should be kept with them.

Sampling kit should contain:

- 3 big plastic or metal cleanable boxes,
 - o one for clean tools and any kind of sample holder or bag,
 - o second for the samples in clean bag,
 - o third for the used, contaminated or just dirty tools.
- markers, tickets for marking the samples
- big sample holders, containers for liquid samples
- plastic bags in different size starting from 1L to 100L depending on the sampled media
- spade, shovel, core sampler, any other kind of sampling tool for soil
- ruler, tape measure for defining the sampled area, distance meter could be useful also for example in a contaminated area

- scissors, knife, secateurs, sickle, lopper, saw or any kind of cutting tool for plants
- protective cloths, disposable gloves, disposable shoe covers, water for washing the tools, paper towels
- personal dose rate meter and contamination monitor if necessary

The instruments and other tools for measurement of radionuclides:

- Gamma spectrometers shielded in the laboratory and portable for field use, preferably high purity Germanium detectors, but scintillation detectors might be useful in case of high load and when measuring just one or two isotope is the interest.
- Low- background alpha-beta counters, mainly proportional counters for measuring gross-alpha and gross-beta, and beta-emitting isotopes in a solid form after chemical separation.
- Liquid scintillation spectrometer for determination of H-3, C-14, gross-alpha, gross-beta from water and so on - sample preparation and separation of the elements is necessary usually.
- Alpha-spectrometers for determination of actinides and lanthanides after chemical separation.

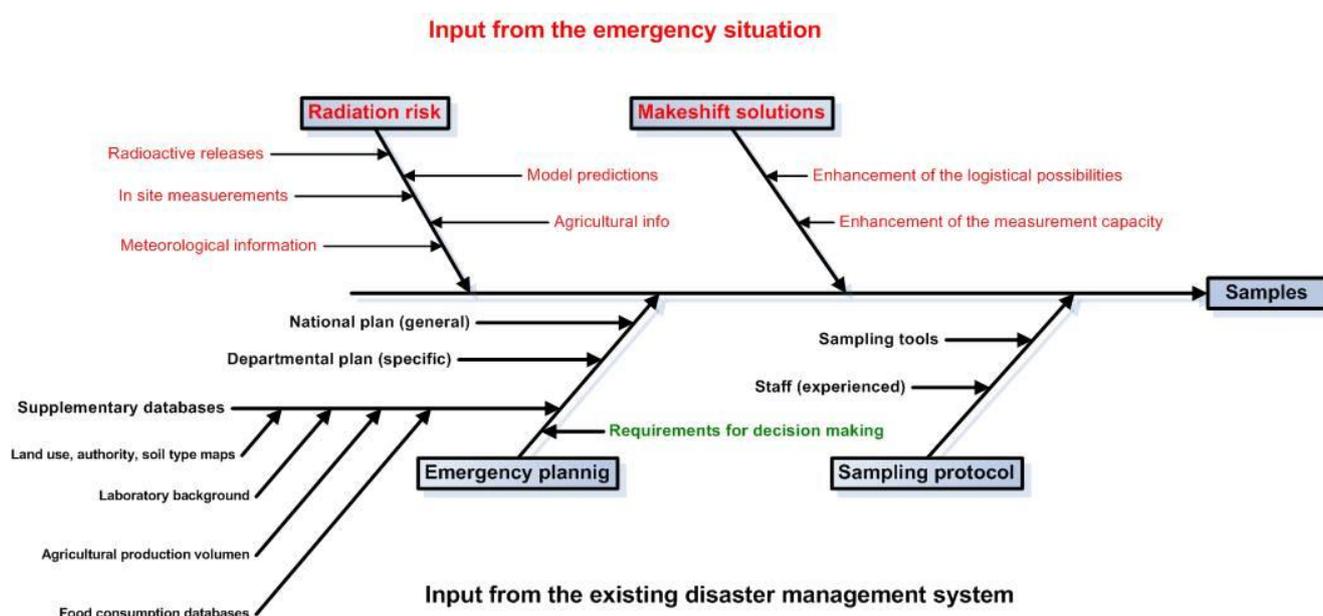


Figure 1: An example of the decision process showing the influencing factors that have to be taken into account

3.0 Case Studies

In the following chapters several case studies are briefly described. All of them are different, all cases have to be treated and studied separately. Decisions need to be made about purpose, approach and process. First purposes have to be identified, then approaches are described, then processes are decided. These case studies can be treated as short summaries with a principal discussion of the presented case at hand that presents novel interest. These studies might be effective for generalizing the possible needs.

3.1 Countrywide Monitoring in Hungary

To develop a countrywide monitoring program thorough and accurate knowledge of the natural, economic, social and sometimes political situation is necessary, or at least necessary to organise a kick of meeting asking all the experts to sit together and discuss the main objectives which are important for the society of the country.

Countrywide monitoring programs always have *multiple objectives* therefore the sampling schedule must be specified even by thinking over by sample type by sample type before the sampling program is designed; the different purposes require different sampling strategies in order to be efficient enough and permit general inferences. In radioecology one eye should be kept on the *natural stochastic variations*, therefore statistical sampling has to be part of the design. Generally the uncertainty associated with the radioanalytical techniques are much less than the natural variation of environmental media. However the representativeness of environmental samples is difficult to demonstrate, samples should provide a true reflection of the targeted population and the environment. *Consistency and comparability* of sampling and measurement raise potential problems with results with different measurement and sampling protocols and with laboratory procedures that might change with time, therefore the harmonisation is necessary.

The main objectives of the countrywide monitoring program are

1. to determine the *radiation dose* to consumers and
2. describe any changes with time to assess temporal trends.

The context of this task involves the *impact of all nuclear related facilities* in Hungary, like the impact of Nuclear Power Plant Paks, all sites belonging to Public Limited Company for Radioactive Waste Management, like Interim Storage of Spent Fuel in Paks, Püspökszilágy Site (low and intermediate level waste repository for smaller radioactive waste producers being out of the fuel cycle, former Radioactive Waste Treatment and Disposal), National Radioactive Waste Repository in Bataapáti (for low and intermediate level of radioactive waste produced in nuclear power plant).

There is no site selected yet for the deep repository of long-lived, high level radioactive waste, however some investigations already carried out and some candidate sites already marked. The program also involves assessment of the impact of nuclear facilities of surrounding countries, those nuclear power plants which agricultural emergency zone cover any part of Hungary. IAEA recommendation is accepted in determination of the zones, namely 300 km circle around the nuclear power plant treated as agricultural zone. There is only a few square kilometres on North-East of the country, which is not necessary to control from this point of view, therefore it is worth to keep the whole territory of the country under control regarding the radioactivity level.

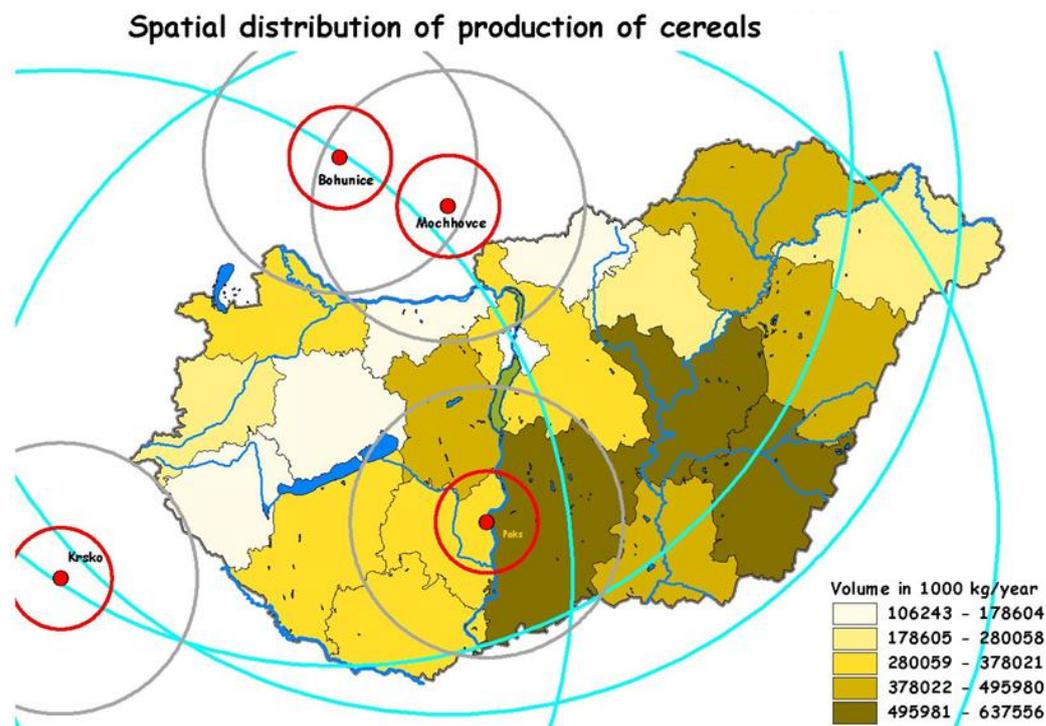


Figure 2: The controlled area for radioactivity in Hungary (red – 30km, grey – 80km, turquoise – 300km zone; last considered as the agricultural emergency planning zone)

The countrywide monitoring programme is able

1. to determine natural and anthropogenic radionuclides to obtain quantitative information on transfer processes,
2. to evaluate present, future and sometimes past situations, and
3. to select the possible short-term or long-term counter-measures,
4. to inform the inhabitants about the actual levels of radioactivity or at least make them being confident that the authorities keep the agricultural environment and food-chain under control
5. to meet the requirements of detecting accidents as quickly as possible, however the monitoring programme is basically focused on long-term, low-level exposures
6. to control the import and export of foodstuffs and feedstuffs.

The sampling strategy has to contain information about the sampling frequency of each kind of sample.

On one hand, there is a need to produce the same type of data every year in order to investigate the long-term tendencies; on the other hand monitoring schedule has to follow the changes in the life of the country, like agricultural production and the actual consumption-structure as well.

The frequency of sampling and measuring the samples was defined to obtain the best representative information about the specific radiological situation. The following aspects were taken into account during the definition of the sampling strategy:

- role of agriculture in the national economy,
- changes in the structure of agriculture and food consumption,

- geographical distribution of production,
- features of territorial units and administrative units,
- organisation of public administration,
- capacity of radiochemical laboratories,
- obtaining a reasonable data set for statistical evaluation.

Knowing well the consumption habits of the population is essential for dose assessment; this has strong effect on the sample types are chosen for monitoring. The recommendations of IAEA Technical Report Series No. 295, 1989 were taken into account. The network should be prepared for answering the questions of the general public, it means that some kinds of foodstuff consumed less than 5% in a year are also involved in the yearly monitoring schedule. Consumption data provided by Hungarian Central Statistical Office come from the evaluation of an appropriate questionnaire. Due to the compromise of needs and the capacity each county has to collect more than hundred samples in a year from different places of the respective area in the frame of the routine monitoring. Regarding the foodstuffs samples are collected from harvested products from open-air production and only the consumable parts of the plants are measured. Hungary has 19 counties (regions); the countrywide sampling schedule is split into county level taking into account the specialities of each county. Other general organisation issue became now regular that the food and feed samples are collected by the officials working in the county offices as “food and feed safety controller” or “plant health officers”; while the samples closely related to the environmental checking of agricultural production are collected by the colleges of the laboratories.

The most important aspects of the sampling schedule are summarized:

1. Milk is collected together with fodder of cows monthly from the same farm year-by-year. Sampling strategy of milk is systematic regarding the time schedule (12 times in a year) and the place also (from same farm), this means on the country level 240 samples including 12 sample-pairs from the farm close to Paks NPP;
2. Milk products: among milk products cheese and milk powder is used in the largest amount in the country. Cheese consumed directly in the households but milk powder not or in very small amount, it is used for wide variety of products in the food industry.
 - a. Cheese: counties having production of cheese have to sample hard or medium hard cheese in the food industry. Others have to take samples from the shops produced in the country if possible. Rather high consumption rate of cheese motivated the double sampling mode for getting information about the production of the country and spatial availability of this foodstuff. The majority of the cheese in Hungary produced from cow milk. Sampling has to be carried out from other type of cheese too, like goat and sheep cheese. The production and the consumption is also much less from these therefore the number of the samples just 20% of the cow cheese. These types of cheese are sampled around the nuclear facilities and where the stocks are high. Sheep and goat are kept in the field not in the stalls in Hungary and the grazing habit of sheep is different, namely they cut the grass closer to the soil than cow therefore the possibility of getting more contaminated cheese is higher. The sampling strategy could be described with stratified random pattern regarding the spatial distribution. The sampling time is determined according to the requirement of having results during the whole year from a region, so the neighbouring counties cover the whole year together. The limited capacity of the measuring laboratories made necessary such kind of monitoring for cheese.

- b. Milk powder is sampled 6 times a year in every county, mainly from producers and from the food industry using milk powder e.g. for baked goods, confections, recombined milk, nutritional beverages, sauces, soups, salad dressings. This sampling can be treated also as stratified random sampling on the country level, because there are two groups according to the place of the sampling: producers and users.

On the country level 240 samples are collected from the milk products in a year. Cheese containing vegetable fat is not target of sampling.

3. Cereals: annually 3 samples are collected from different locations in the county, every county has production from them, just the amount and kinds are different – cereals considered basic foodstuff in Hungary, the number of samples are 57 and the highest portion is always wheat as in the production and consumption, but there are some rye, barley and oat samples too. Sampling of cereals are treated in the country level as random sampling regarding the spatial distribution, because no instructions are given for the sampling place; timing is determined by the harvesting period;
4. Meat: 6 beef, 6 pork, 5 poultry samples are collected in each county. Sheep, rabbit and other type of meet collected just from the grazing area in smaller numbers. Fish from fresh water lakes or rivers is collected once a year; however, the consumption rate is low, 1.6 kg/year per person, there was a request from the public to control it. Meat of wild animals has to be collected from the official collecting places and taking into account of good hunting areas of the country, so finally meat of wild animals are collected from half of the counties, and the portion of these samples is 15% from the total of 390 meat samples in a year and the timing of collection is determined by the hunting seasons. For grazed animals sampling is evenly distributed in a year for two reason, first of all sample preparation and measurement capacity required this and of course for having some temporally distributed data too. For domestic animals the temporal distribution has not big importance, because they are kept in stalls. There was a need to clarify that the sample amount collected has to be understood as the weight of the muscle;
5. Vegetables have to be collected mainly from open air production and vegetation period has to be taken into account in timing of the sampling.
 - a. potato: it has important role in Hungarian cuisine, samples has to be collected from the production of the current year and samples has to be treated after the washing and without peeling them;
 - b. root vegetables: warning are the same as in case of potato samples;
 - c. leafy vegetables like spinach, sorrel, lettuce are sampled 2 times (end of spring and autumn) 3-3 samples from different places of the county – leafy vegetables produced open air are good differential bioindicators, showing well the short term changes having large adsorbing surfaces therefore worth to deal with them in the monitoring program ;
 - d. other vegetables also has to be sampled, like tomato, green paprika, green peas and beans, onion, garlic, maize, different kinds of cabbages having longer vegetation period than leafy vegetables – those are consumed in bigger amount in the country;

Sampling of vegetables treated as random sampling having production in the whole country starting from the small gardens to a big farms, no additional special requirement is defined; 330 samples collected from vegetables in a year from the whole country.

6. Fruits: apple has the highest production and consumption rate, 4 samples are taken in a year and 1 sample is gathered from different kind of berries including grape. The total number of the samples are 100 on the country level and the type of the sampling also random, reason is the same as in case of vegetables;
7. One tenth of the samples are so called freely chosen samples according to the specialities in production of the county, like red paprika as a powder or additional samples from sheep cheese – the types are usually agreed and harmonised at the beginning of the year, but the collection of them is not so strictly checked, they might be substituted according the yearly production or the influence of having the extreme weather conditions;

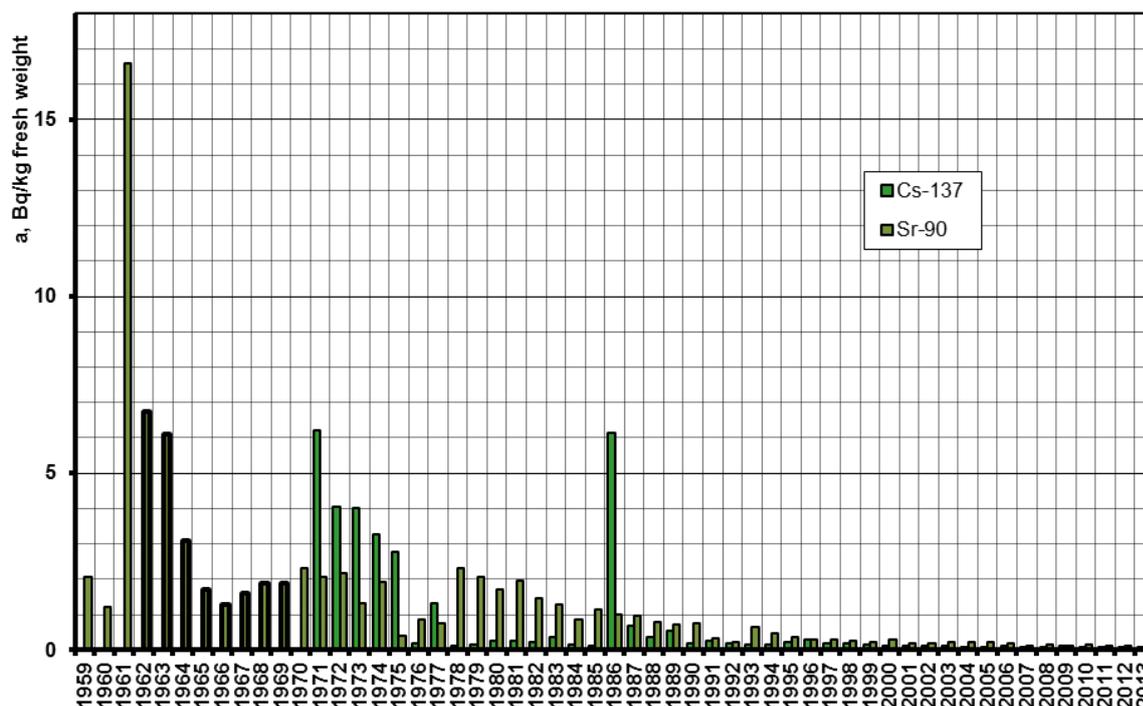


Figure 3: Long-term countrywide averages of leafy vegetables sampled in Hungary as an example of the use of the monitoring data (data before 70's were calculated from recent measurement from several preserved samples)

Sampling schedule for the laboratories working in the food industry is of course individually determined just giving some advice for them. Until first half of 2000's analyses of cereals, milk products, meat and canned foods gave a very good additional database for the country data.

Collecting pair of samples from the same location is important from the ecological point of view; deriving the transfer factors giving the importance of this kind of sampling and the most important use of the locally valid transfer factors is to predict the contamination and the ingestion dose of inhabitants in an emergency situation, like

- leafy vegetable - soil
- milk - fodder,
- grass - soil, alfalfa – soil.

Monitoring → transfer

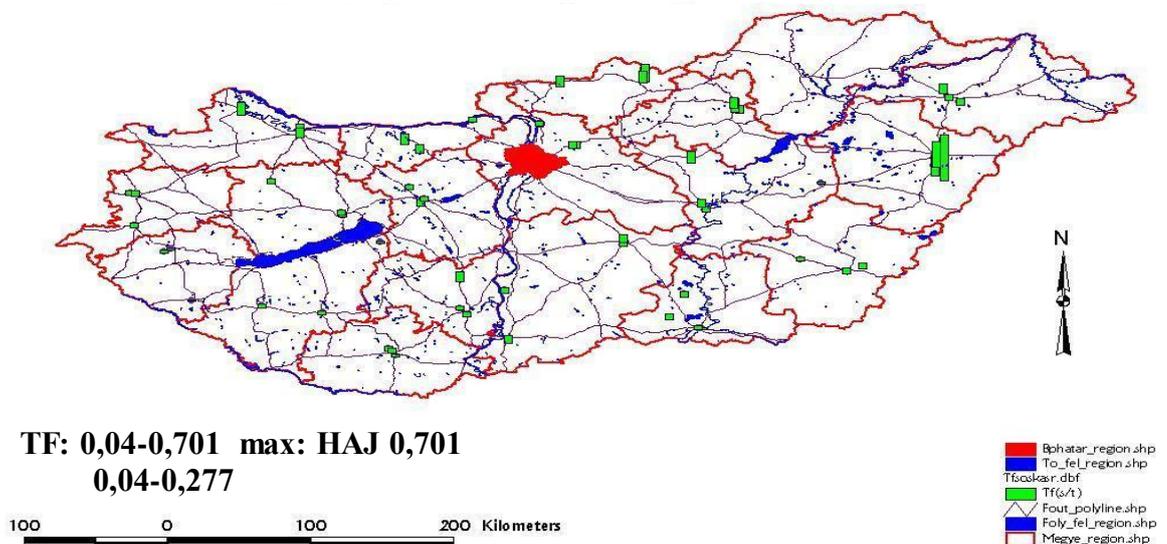


Figure 4: Spatial distribution of transfer factor (grass/soil) of ^{90}Sr , derived from 10 years database from sample pairs

The network collects samples, controlling the semi-natural and natural environment as well, like meat of wild animals, moss, and mushroom picked up in a forest or grazing land. Some of them considered as bioindicators, they are also involved in normal sampling programme to get information during the whole year. In this way the network is able to provide data about the activity concentration of different radionuclides in different medium, time dependency and spatial distribution of radionuclides in soil, plants, foods and feeds, in order to give base to issue export allowances, to assess ingestion doses and to give support to decision-makers in the case of an incident or an accident at a nuclear facility.

1. Grass and soil sample pairs are commonly collected in a monitoring program, during the sampling worth to measure and note the area from where the sample was collected for calculating the fall-out in the unit of Bq/m^2 and the cutting height might be also important, cutting in 0.5 cm height or less is the habit of sheep type animals, 2 cm gives information for food chain of the cow types. The other sampling advice might be to collect soil samples from the same area at least from the upper 5 cm layer. The most informative is the core sample for soil from an undisturbed area. The usual slices of the core are 1 cm or 2 cm sometimes 5 cm, but not thicker, the recommended depth is 30-50 cm, depending the thickness of the soil in the field. It is worth to collect the soil from the same surface as the grass. From these dataset the transfer factor between the grass and soil can be calculated, the inventory from the fall-out can be determined and if one part of the grass sample is washed and another not, the rate of the resuspension can be estimated as well. If this rate is not negligible it is better to calculate the transfer factor from the result of the washed grass.

Table 1: Radioecological importance and sampling frequency of environmental media influencing the agricultural production in a low dose environment, when the goal is to determine the long term changes

Sampled media	Properties, influence, achieved goal collecting them	Sampling frequency
soil	works as reservoir of radionuclides, long term effect on the agricultural production, continuous effect during the production period	once a year from same place
moss, lichen	integral bioindicators effectively collecting radionuclides from the fall-out during a long period, well applicable for determination of spatial distribution of contamination	once a year from the same area
wild mushroom	bioindicator collecting radionuclide from a large area through the net of mycelium, there is some delay appearing in their activity concentration if fresh fall-out occurs (there are some specificities, like Boletus collects especially well Ag isotopes)	very dependent on the vegetation period, 3 times in a year having attention on the species
plants, vegetables with big leaves	differential bioindicator, immediately shows the fresh fall-out (be careful in dry period, if the plant grows on the contaminated land, there might be the resuspension of the soil)	monthly in the vegetation period
rain	“wet fall-out” might bring down 10 times higher or even more activity than dry	continuous sampling with a period of a month, if nothing happened in any nuclear facility, the period has to be shortened if an event occurred taking into account the model predictions and measurement capabilities
aerosol	immediately indicates the contamination from fall-out	continuous sampling with a period of a week if nothing happened in any nuclear facility, the period has to be shortened if an event occurred taking into account the model predictions and measurement capabilities

	January	February	March	April	May	June	July	August	September	October	November	December	
Cereals	passive			growing		harvesting			sowing	passive			
Potato				sowing	growing			harvesting					
Root vegetables				sowing	growing		harvesting						
Onion, garlic			sowing	growing		harvesting							
Tomato				sowing	growing			harvesting					
Green paprika				sowing		growing		harvesting					
Cucumber				sowing	growing	harvesting							
Green peas			sowing	growing		harvesting							
Green beans				sowing	growing		harvesting						
Cabbages			sowing	growing				harvesting					
Lettuce			sowing	growing	harvesting								
Sorrel	passive		sowing	growing	harvesting			passive					
Spinach	passive			sowing	growing	harvesting			passive				
Apple	passive					growing			harvesting		passive		
Peach	passive					growing		harvesting		passive			
Grape	passive						growing		harvesting		passive		
Plum	passive					growing			harvesting	passive			
Sour cherry	passive				growing	harvesting		passive					
Cherry	passive				growing	harvesting	passive						
Pears	passive					growing			harvesting		passive		
Raspberries,	passive				growing	harvesting		passive					
Strawberry						sowing	harvesting						
Mushrooms	passive						collecting					passive	
Forest berries	passive				growing	collecting			passive				
Fresh fodder,	passive				growing	harvesting					passive		
Maize				sowing	growing				harvesting				
Sunflower			sowing	growing			harvesting						
Honey					collecting								

Figure 5: Typical vegetation period of agricultural products in Hungary (white – no effect expected, no part of the plant is on the field)

Some practical aspects for defining the sampling strategy and executing the sampling:

1. Aerosol sampling is continuous mainly with one week collection period: preferably large volume aerosol filter (> 40000m³/week) with large surface (> 0.25m²). The flow rate and the thickness of the aerosol filter should be adjusted to achieve almost no pressure difference in the incoming and outgoing air of the filter. For large filter a wired frame might be applied for providing the stability of the filter material. After the collection period the sample has to be folded or pressed keeping inside the exposed side of the filter. A standard geometry has to be applied for the measurement either pressed or just folded.
2. Precipitation or fall-out sampling is continuous mainly with 1 month collection period: tool is simply a 1m by 1m stainless steel tray with a possibility to collect the precipitation into a 10-20L plastic container with small mouth (for prevention of the evaporation losses) for example from the outlet of a corner of the tray. This needs a careful watching the amount of precipitation and changing the container when necessary.
3. Soil sampling might be from a given area or from the spot defined in the monitoring program. The minimum number of the samples from “homogenous area” in some environmental features has to be defined as

$$s = \sqrt[3]{n},$$

where

- s minimal number of the sampled units
- n total number of the units on the field.

The amount of sample collected from one unit depends on the sample preparation protocol and the applied measurement protocol requirements (like geometry: Marinelli baker with volume of 1L or e.g. 0.1L cylindrical metal container; necessary amount for chemical separation ...).

When the survey of an area is the goal sampling strategy might be

- a. systematic sampling – usually applied on a field where there is no extremity, no change in the geological formation, might be ideal for a plain;
- b. random sampling – might be applied in the same case that mentioned in 3.a. or for example when the environmental characteristics of the field cannot or hard to determine well, for example in a forest containing many kinds of trees;
- c. stratified random – generally this method applied because the most area has some characteristic which has to be taken into account, usually this case means that the area might be divided for several parts differing in environmental characteristics, for example there is a river or a pond in the area, random sampling might be executed along the river or at the bank of the pond and random sampling in the “terrestrial area”. Such differentiation might be inside the forest if there are tree groups from same species in some parts of the forests – it might be interesting to sample each kind of sub-forest.

The goal of the soil sampling defines the tools have to be used, there might be some special cases, but the general guidance is the following:

- when the target nuclides are actinides or lanthanides or even a fresh fall-out just the upper 2cm has to be sampled – shovel or any other tool is sufficient for collecting or scrubbing just the upper layer of the soil;
- in an agricultural area where the soil is well cultivated a simple spade is good for sampling if those properties of the soil has to be determined which has effect for the plants root uptake. In this case worth to get information in advance that soil how deeply is cultivated year by year;

- in an undisturbed area worth to use any kind of core sampler to define the depth distribution of the radionuclide of interest. Worth to apply changeable inner tube for easier executing and keeping the samples in these tubes until in the laboratory they are sliced into the proper layers – 1cm, 2cm or 5cm layers are applicable depending of the soil properties. The top of the soil sample tube always has to be marked on the field.
4. Water samples might be any kind of surface water, groundwater, tap water, mineral water, water from a household well or well for food industry
 5. Biological samples are covered by food and feed and bioindicators in Hungary. All of them have a bit different goal and aspects. Generally the sampling strategy for bioindicators is random sampling inside the county, of course the samples can be collected where they are encountered (from a sandy area hard to collect moss samples for example), but Hungary being rather small country there are not big differences in the environmental conditions. For vegetable sampling the random strategy fits well

Knowing the general level of anthropogenic isotopes inside the country the sample amount is defined by the achievable minimum detectable activity concentration in material of interest. Nowadays in Hungary concentration of anthropogenic isotopes are low. Taken into account the requirements laid down in 2000/473/EURATOM [3] directive regarding the ability of measuring the actual level of radioactivity the following amount is necessary to collect from different kinds of media:

Table 2: Sample amounts of foodstuffs and feedstuffs necessary to collect for measuring the actual level of anthropogenic radionuclides in Hungary

Sample type	Sample amount in fresh weight, kg	Remarks
Moss	2	Collection from flat place, like big stone, roof of the house – bigger than 1m ² continuous surface covered by moss is ideal
Mushroom	4	Preferably edible mushroom from one kind or at least from same family
Milk	5	Preferably from a local farm sampled together with feedstuff of animal
Foodstuffs (fruits, vegetables), herbs (nettle, camomile...)	4	Local products with well definable location of harvesting
Different kinds of corns either for human or animal consumption	1.5	Local products with well definable location of harvesting
Fresh feedstuffs, pasture, alfalfa ...	3	From 1 m ² or bigger area cut in 2 cm height, preferably without soil
Dried foodstuffs, feedstuffs, mixtures, additives	0.5	Preferably local products, in case of additives at least the country of origin should be determined
Samples of animal origin with high water content either foodstuffs or any other (meat,)	3	Local products with well definable location of living or kept for domestic use

Sample type	Sample amount in fresh weight, kg	Remarks
Samples of animal origin with low water content either foodstuffs or any other (bones, claw, horn, antler)	1	Local products with well definable location of living or kept for domestic use
Controlling import foodstuffs and feedstuffs	1	From third county (term of European Union – outside of the EU), the limit is high enough to proof from smaller amount of sample

Some general requirements has to be kept like

- foodstuffs samples has to fulfil the general consumption requirements and has to be take care about the reservation until arriving into the laboratory;
- the samples have to be marked in a well identifiable way;
- Sampling report has to contain the location, date of sampling, name of the person taken the sample.

Sampling report always has to kept and delivered together with the sample.

3.2 Environmental Impact Assessment of the Incident of Nuclear Power Plant Paks in 2003

Several fuel assemblies were severely damaged during a fuel crud removal operation on the Paks-2 unit on 10 April 2003. The assemblies were cleaned under the water in a deep tank, the most of 30 fuel assemblies suffered heavy oxidation and fragmentation because the inappropriate cooling; the first sign of failures was the detection of fission gases. The event was reported to IAEA and classified as Level 3, severe incident on the International Nuclear Event Scale (INES).

Table 3: Activity release from damaged fuel during the incident [4], [5]

Isotope	Released activity, Bq	Relative release, %
⁸⁵ Kr	6.87E+14	3.00
¹³³ Xe	6.6E+14	1.19
¹³¹ I	5.9E+14	1.41
¹⁰⁶ Ru	8.7E+12	0.03
¹³⁴ Cs	4.2E+13	0.74
¹³⁷ Cs	3.8E+13	0.53
¹⁴⁰ Ba	1.8E+14	0.16
¹⁴⁴ Ce	7.2E+13	0.06

The incident happened 2 weeks after the shutdown of the reactor, therefore there were no isotopes with very short half-life in the system. The isotope specific data of volatile gases released were determined with a continuous monitoring system of NPP, the release values were recorded in every 10 minutes. The highest release was detected for ¹³³Xe with 10¹³Bq/10min, ⁸⁵Kr with 10¹²Bq/10min, ¹³¹I with 10⁹Bq/10min and 99% of the total release took place in 24 hours. The monitoring system of the release provided information about the

form of iodine, 77% of the total ^{131}I release was in form of elemental iodine, 21% was organic and 2% aerosol [5].

The NPP has regular environmental monitoring program around the facility inside 30km circle. There are stations controlling only dose rate but there are some automatic (A type) measurement systems controlling aerosol, iodine isotopes (different forms) ^3H , ^{14}C content of the air and stations controlling the liquid release too. According to the dose rate measurements of control stations around the facility and taking into account the weather conditions there was any special countermeasures suggested. The highest dose was measured in the A type control station being in the actual wind direction, 421nSv/h which went back to the normal background level of 70nSv/h in an hour. The alarm level is set to 500nSv/h in Hungary.

The highest value of measured was 13Bqh/m³, while the calculated was 27Bqh/m³ as highest.

For assessment of the environmental effect of the incident the release data of the NPP were available – as they are listed in the Table 3. Among those volatile isotopes the ^{131}I , ^{134}Cs and ^{137}Cs have environmental impact, therefore monitoring strategy were elaborated taking into account the properties of these isotopes only. The other important information came from the Hungarian Meteorological Service, they provided the wind trajectories for every 3 hours starting with the time of the incident. Just a remark that the main wind direction is North-West in Hungary, but at the time of the incident the wind direction was South then it swiped almost the whole Plain of the country in the next few days. So the place of the sampling had to be appointed according to these changing wind directions. This territory has the highest importance in the agricultural production. The state of the agricultural production had to be taken into account, while the incident took place at the beginning of April, i.e. at the very beginning of the vegetation period, as even Figure 5 shows, the cereals had just 4-6cm green parts.

Before starting the monitoring there was a need to assure that the sampling team does not need any special protection, therefore several in situ measurements were performed by HPGe detectors. In the measurement campaign four mobile laboratories took part. On the map of Figure 6 the results are summarized, the highest measured deposition was 360Bq/m² for ^{131}I . The other isotopes were not detectable even in close vicinity of the NPP. Table 4 shows the detailed results of Department of Radiochemistry of National Food Investigation Institute (former National Food Chain Safety Office).

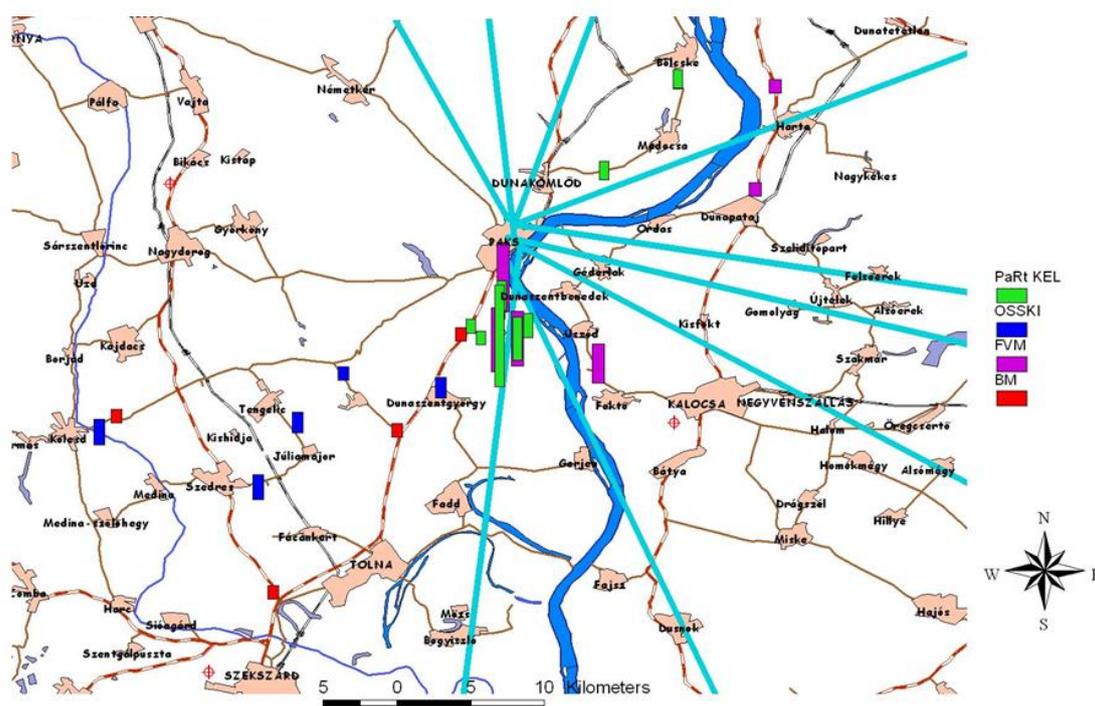


Figure 6: In situ measurements by HPGe detectors right after the incident. The highest deposition results was 360Bq/m^2 of ^{131}I measured in 3km distance from NPP (height of the colour columns are proportional with the results of the measurements; turquoise lines are wind trajectories)

Table 4: Detailed data of in situ measurement of anthropogenic radionuclides performed by mobile laboratory belonging to Ministry of Agriculture – only ^{131}I was fresh fall-out.

Settlement	Latitude	Longitude	a, ^{131}I Bq/m ²	a, ^{137}Cs Bq/m ²
Solt	46.79585	18.96231		962
Dunapataj	46.65596	19.00723		1539
Uzód	46.54929	18.91158	139	543
Harta	46.71924	19.01956		1099
at K-5 station	46.56401	18.85012	227	531
at A-4 station	46.56449	18.86255	197	646
NPP (North entrance)	46.60217	18.85367	247	764

There was no other possibility than check the contamination level of grass-soil or young cereals-soil sample pairs. Stages of the planning:

1. Objective: assessing the effect of the incident
2. Identification of the target: measuring only those isotopes which might have environmental effect in early spring, like ^{131}I , ^{134}Cs , ^{137}Cs , all of them might be determined by gamma ray spectrometry.

3. Target population: grass, nettle or young cereals and soil from the same place, sample pairs are sufficient (no need core samples, because of the fresh fall-out).
4. Sampling design: random sampling, sampling area defined day by day according to the changing wind trajectories.
5. Data necessary to note during the sampling:
 - a. date and time,
 - b. geographical coordinates of sampling place,
 - c. dose rate,
 - d. surface of the plant sampling
 - e. surface of the soil sampling,
 - f. weight of the sample – weighing on the field while the samples put into Marinelli beakers there.

At that time there was no information available about the dose rates measured around the NPP. The first sampling was on 11 April and it continued until July 2003. On the Figure 7 the 2 most important sampling tools are shown. The applied measurement technique was gamma-ray spectrometry in laboratory conditions, Marinelli beaker with 450mL volume was used and 10,000s measuring time was sufficient enough for the 30% relative efficiency HPGe detectors in a 10cm lead shielding.



Figure 7. The most important tools on the field were Marinelli beakers for samples and GPS to mark the locations.

The results are summarised on the Figure 8. In April and May 121 samples were analysed in Radioanalytical Network of Ministry of Agriculture. The collected samples were mainly grass, 79% and young fresh green wheat was 13%, the other were nettle and alfalfa. 12 samples were below detection limit, the majority of the samples were between 1 and 30Bq/kg and there were 4 samples between 200 and 500Bq/kg and only one above 500Bq/kg. The release of the first minutes resulted the highest values North from NPP, but mainly in 3km distance.

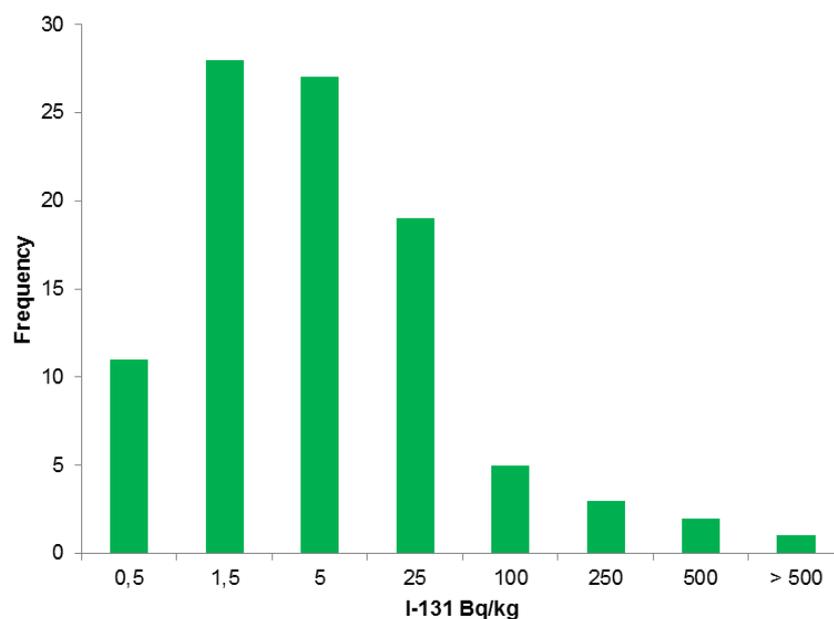


Figure 8: ^{131}I results measured in green samples (grass, nettle, cereals) during April and May 2003.

The Figure 9 shows the spatial distribution of the ^{131}I contamination after the incident. The deepness of the colour is proportional with the activity-concentration of green samples measured in fresh state. All of the six results above 200Bq/kg activity concentration were inside 3km circle around the NPP and was the result of the first “big” plum at the very beginning of the incident. The locations of those data allowed to draw this conclusion and the wind directions proved this theory. The results were decay corrected to the day of the incident.

Because of the quickly changing wind direction there was no time to elaborate a systematic grid of sampling and it would not be efficient enough, because the distribution of the fallout was not evenly. The sampling points were determined day by day according to the information from NPP and from Meteorological Service.

The estimated dose for public was 130nSv as the most conservative value taking into account three main ways of the exposure like

- external exposure,
- inhalation and
- ingestion doses.

The annual dose constraint for NPP is 90microSv. The incident seemed a good real time exercise for the monitoring network and last but not least there was a good trial for checking the efficiency of the communication between organisations belonging to different ministries and communication to public.

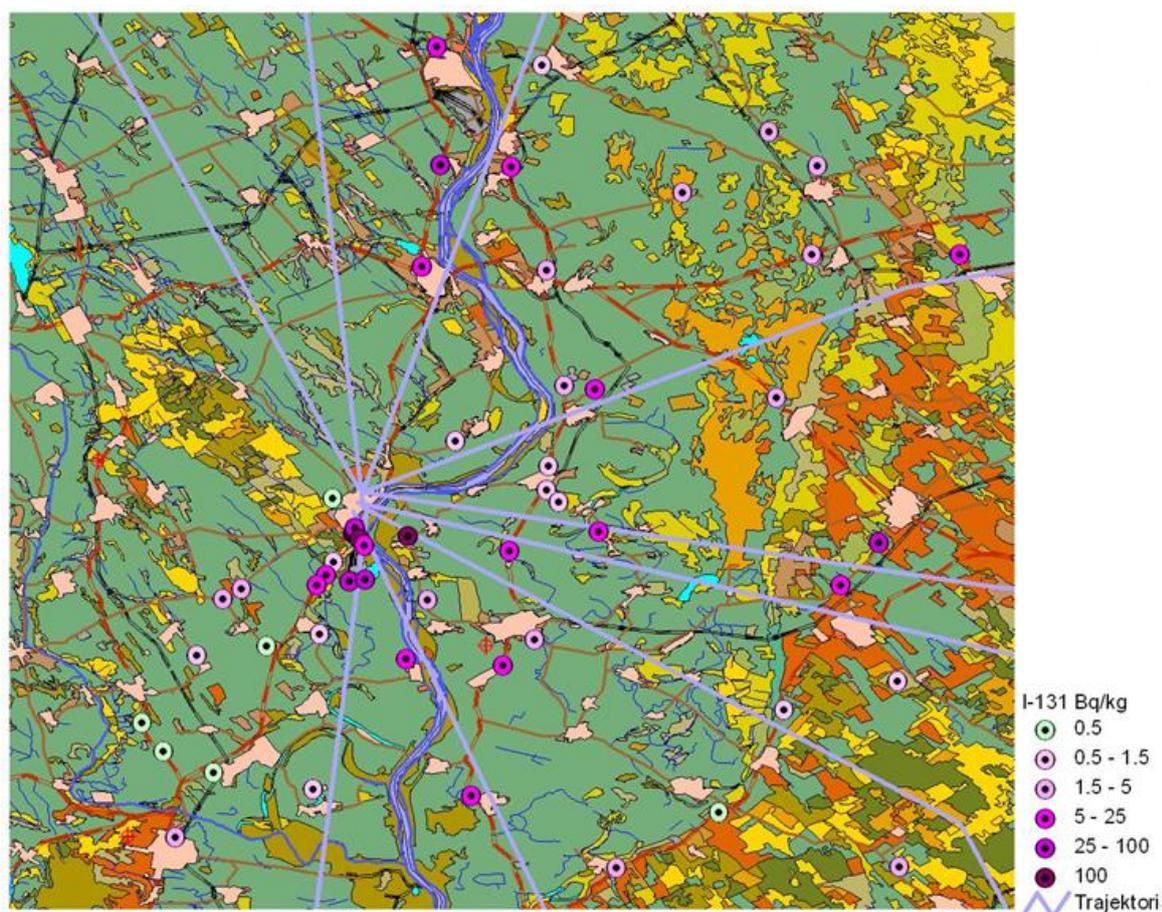


Figure 9: Sampling points and measured activity concentrations of ^{131}I in the vicinity of the NPP

3.3 Monitoring the effect of the accident of Fukushima Dai-ichi Nuclear Power Plant in Hungary, 2011

After 11 March 2011 a big earthquake (grade M9) followed by a gigantic tsunami with the waves of more than 10m a serious accident occurred in Fukushima Dai-ichi Nuclear Power Plant, it was classified as serious accident, 6th grade on the International Nuclear and Radiological Event Scale (INES).

In Hungary different institutions and different networks were available at that time for measuring radioactivity in aerosol samples. The purpose of the installation of the aerosol samplers were different, the majority of them is able to detect the effect of an accident but some of them is designed for giving adequate information even in case of severe incident, i.e. INES 3 or even INES 2. The performance characteristics of these units are totally different, just mentioning only 3 types among them:

1. sampler with small filter surface approximately 20cm² and 0.5m³/h airflow,
2. sampler with a medium surface of 500cm² and 140m³/h airflow,
3. sampler with 2700cm² surface and 230m³/h airflow.

Without harmonisation of the sampling methods and evaluation requirements the institutions belonging to different organisations and ministries reported the values shown on the Figure 10. For the better understanding even for the public or for the decision makers there is an essential need to report the uncertainties of the measurement values – without them one can have a feeling about the order of magnitude of the contamination, which might be very useful

in case of a severe accident when the decision has to be taken very-very quickly. This level is not enough when the goal is the estimation of the long term consequences of an incident or accident. The higher capacity sampler is able to give the results with sufficient relative uncertainty of less than 10%, or even less than 5%. This confidence is good enough for the planning of the agricultural countermeasures and for the input data for predicting the possible contamination levels in the food chain. Institutions gave the approximate detection limits without any comment – if the evaluation is done according these values all of them are reliable. If there is a need to have 99% confidence, better to apply the worst case and consider the given detection limits as signal to noise ratio converted to actual measurement unit, only 11 results from 19 are acceptable, but still without uncertainties. These are written with normal letters on the figure and values with italic numbers are below this confidence level.

Measurement results given with uncertainties were provided by Central Agricultural Office and the data are detailed in Table 5, the collection time was a week and after 3 days cooling time the measurement was performed by HPGe n-type detectors with 30% relative efficiency mounted in ultra-low background cryostat in a low background counting room with 80000s measuring time. From the results is obvious the role of the time elapsed between the accident and the observation, because in the second period ^{132}I was not detectable at all. $^{110\text{m}}\text{Ag}$ is an activation product, it was interesting to discover it in the spectrum. One eye is kept always on the natural isotopes, because their values serve as inner quality control of a sample. One more remark about the given data – for the raw database of the network combined uncertainties are collected without applying any coverage factors (extended uncertainty) for avoiding misunderstandings later on.

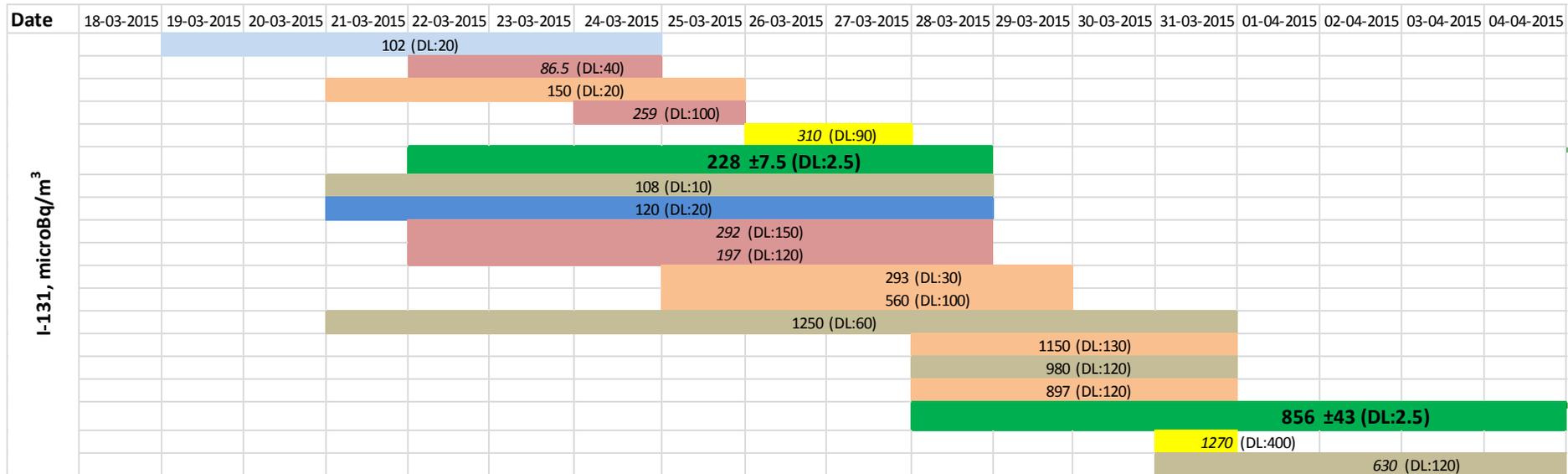


Figure 10: The activity concentration in aerosol samples in Hungary. Data were collected from different institutions having aerosol samplers for different purposes. Aerosol samplers have different designs, collection times, sucked air volumes, measuring instruments differs from each other. The bars indicating the sampling periods, colours are indicating the same network of units but not the same geographical places and the style of the numbers are indicating the reliability of the data, namely: italic – values below the confidential detecting; normal – above the confidential detecting; bold – reliable results, the measurements having 2 data are the results with uncertainties, e.g. $228 \pm 7.5 \text{ microBq/m}^3$ and $856 \pm 43 \text{ microBq/m}^3$

The accident had negligible impact on the Hungarian environment and population because of the low level of the contamination and the short “exposition period”. The elevated level was measurable just from short lived radionuclides. According to the official opinion of the health authority there was no health risk to the population due to the radiation danger – all the data listed here are proving the statement.

Table 5: Detailed measurement results of the most precise measurements

Nuclide	22-28 March 2011, air volume 32848m ³		28 March – 4 April 2011, air volume 38651m ³		Detection limit, micro Bq/m ³	Identified gamma-lines, keV
	Activity concentration, micro Bq/m ³	Uncertainty (k=1), micro Bq/m ³	Activity concentration, micro Bq/m ³	Uncertainty (k=1), micro Bq/m ³		
^{110m} Ag	2.18	0.4	-	-	-	657.75; 884.67
¹³¹ I	228	7.5	856	43	2.5	364.48; 284.3; 636.97
¹³² Te	8.8	2.3	18.7	4.3	2.5	228.16
¹³² I	8.9	1.6	-	-	5	667.69; 772.6; 954.55
¹³⁴ Cs	12.0	1.4	56.3	3.1	1.5	604.69; 795.84; 569.32;
¹³⁷ Cs	11.6	1.5	58.7	3.8	1.5	661.67
Natural radionuclides						
⁷ Be	4350	130	4357	216	10	477.59
²¹⁰ Pb	405	24	547	45	24	46.51

3.4 Strategy for Finding Proper Reference Material in Fukushima Prefecture

The study was performed in 2014 in the Institute of Environmental Radioactivity of the Fukushima University. The authors believe that there is a need for forming an institute which is able to offer a help for public and private institution willing to deal with the measurement of radioactivity in the Prefecture or even on the country level with expertise and with providing well characterised in-house reference materials in a well usable quantity.

The reliability of the measurement results is essential to convince the public of the measures taken by authorities are perfectly prepared and established. A well-maintained and consistent database is the crucial point of the monitoring and reference materials are important tools in quality management in the laboratory and on the network level too. Reference materials intended to prove the reliability of measurements therefore they might have some role in the local society too, like to achieve the public acceptance of the results and to eliminate the fear without any problem and further consideration.

By the definition, reference material is “sufficiently homogeneous and stable with reference to specified properties, which has been established to be fit for its intended use in measurement or in examination of nominal properties.” Providing the associated uncertainty with reference values is also required. Reference material can be used for measurement precision control

whereas only reference materials with assigned quantity values can be used for calibration or measurement trueness control. Reference materials have to be well characterized and homogeneous for a large number of analytes; therefore, they are useful in:

- method validation,
- method development,
- long-term performance test of the method (Shewhart and cumulative sum control chart),
- defining several properties of a method, like repeatability, reproducibility, limit of quantification,
- assessment and quantification of different sources of uncertainty taking into account uncertainty of the reference material,
- estimation of accuracy and precision,
- evaluation of possible interferences.

Recently one certified reference material is available in Japan produced in the Fukushima Prefecture and prepared in collaboration of the National Metrology Institute of Japan, The Graduate University for Advanced Studies and National Food Research Institute [6]. They prepared brown rice put into U8 container which is generally used for the measurements performed by gamma spectrometry. This material has importance from two reasons, on one hand the activity concentration is less than the legal limit of consumption, but in the same order of magnitude and on the other hand they prepared rice, the most important food in Japan.

Before going the field and collecting samples for reference material there are a couple of things to think over thoroughly. First of all, the stages of the planning of a pilot study are the following:

1. Objective: finding a proper reference material for radioanalytical purposes; primary goal was to use them for gamma ray spectrometry therefore having three reference material with different densities and different levels of radioactivity would be useful.
2. Identification of the target:
 - a. radionuclides: ^{134}Cs , ^{137}Cs were obvious, natural isotopes, gross-alpha, gross-beta and ^{90}Sr .
 - b. material: minimum number of required units are 50 and maximum 200; unit size is determined by the Japanese common use, so-called U-8, which is a 100mL plastic container. The necessary amount of the reference material derived from these data and the required densities, which are
 - i. 0.3g/cm^3 – represents dry plants,
 - ii. 1g/cm^3 – represents any of fresh foodstuffs and feedstuffs (plants, milk, meat...)
 - iii. 1.6g/cm^3 – represents the highest density of soil and sediment.
 - c. Target population:
 - i. dry plants: in the environment of Fukushima smaller or biggest bamboo forest or just spots available everywhere. The use of bamboo is general even now, therefore the public acceptance of this kind of reference material might be foreseen.
 - ii. fresh foodstuffs and feedstuffs: because of the requirement of long shelf-life it is not possible to use natural material for this purpose. The recommended material either the purified water or preserved gel spiked

with the required radionuclides. Generally preservation of natural water also might be good, but still remains a risk of deterioration.

- iii. dried soil or sediment is sufficient.
3. Sampling design: there are available studies about the contamination of area, either dose rate maps, surface contamination maps, or measured activity concentrations in the different media. These maps are freely available on the webpage: <http://ramap.jmc.or.jp/map>. From the studies already conducted there is another important finding that the dose rate is fairly proportional with the concentration of radiocaesium in soil (concentration of ^{134}Cs and ^{137}Cs together).
4. Data necessary to note during the sampling:
 - a. Code of the sample - a consequent coding has to be established which might be used for decades for the easy tracking back the materials,
 - b. date and time,
 - c. geographical coordinates of sampling place,
 - d. dose rate,
 - e. weight of the sample,
 - f. estimation about how big is the amount that might be collected from the same place.
5. During the pilot study the loss rate i.e. the portion of the useless and final product has to be determined for collecting the correct amount of the material. Losses might occur in any stages of the sample preparation starting with drying, ashing, grinding through the separation steps like sieving until putting the material into the proper packing. The consumption of any kind of testing among them the homogeneity test and the characterisation has to be considered as well.

One of the most important decisions has to be made about the preservation of the candidate material, because the stability of the reference material is essential for the long-term reliable use. There are two types of requirements in this context; short term stability has to be proved modelling the possible transportation circumstances e.g. thermal stability of the material has to be proved practically between -50°C and $+70^{\circ}\text{C}$ [7], so the certified property of the material will not change. The long-term stability concerns the stability of the property values under shelf life circumstances for a long period. At least a reference temperature has to be chosen, and sometimes the optimal humidity also has to be given. Many biological and environmental candidate reference material show some instability despite the effort of determination optimal storage conditions. Usually dried or ashed materials are stable enough to keep and use them in normal circumstances, e.g. at 20°C and less than 30% humidity through several years. Never forget about using rather narrow range of particle size for preventing the simply physical segregation process. The obligatory irradiation might be also useful even for this purpose.

The explanation of the chosen densities is the following:

- $0.3\text{g}/\text{cm}^3$ – there are many kinds of dry and instant food are available in shops of different chains of grocery stores, like the enormous variety of cup noodles, instant ramen, instant lunch, miso cups, instant yakisoba noodles; amazing that even sushi and tempura can be enjoyed as instant food in Japan. There are many kinds of dried seafood available, a lot of sauces are sold also in instant forms and so on. The density of dry bamboo leaves match with the density of these products, therefore the direct comparison is also possible.
- $1\text{ g}/\text{cm}^3$ – any kind of biological sample in fresh usually has density around this, moreover the water samples either fresh or sea water also might be directly compared. The shelf life is the problematic point again, the easiest matrix would be the water

sample, but reservation and the wall effect of the bottles stored in might be the crucial and sometimes not resolvable problems.

- g/cm^3 - usually the upper value of the soil and sediment. Any of them may be properly prepared for reference material, there is no need to think of the similar densities.

The pilot study was carried out at the beginning of 2014. During the winter period the sampling possibilities were determined by the local climate and weather. The goal was to prepare a special, local reference material; Japanese love trees and respect their traditions and the fact that in any part of Fukushima Prefecture is possible to find smaller or bigger spots of bamboo brought the idea to have their leaves as reference material. Somehow the acceptance of the reference material should be achieved. There are two more big advantages of this plant that it is perennial evergreen plant and it grows in small groves rather than in extensive contiguous areas everywhere in the country. Bamboos are some of the fastest-growing plants in the world due to a unique rhizome-dependent system. Bamboos are of notable economic and cultural significance in Japan but mainly in South-Southeast-East Asia, being used for amazing variety e.g. food source, and as a versatile raw product. High-quality bamboo is stronger than steel, a property that has made it a choice even in weaponry or building materials. The importance of this plant is shown even in the Statistical Yearbook of Japanese Statistics Bureau they have the following big categories: Total, Artificial forest, Natural forest, Treeless land and Area of bamboo, however it is just 159,000ha, 0.6% of the total area of forests. Privately owned portion is also remarkably, 94% and all of them fall into non-national forest category in Japan [8]. Leaves can be freely collected anywhere in the Prefecture, because there is no special importance of this part of bamboo for the owners.

The amount of the final reference material is detailed in the Table 6. From the experience gained in the field of preparation of matrix reference material in the National Food Chain Safety Office as the collaboration centre of IAEA, generally might be stated, that the sampled amount has to be at least three times as the final, treated (mainly physical) and homogenised candidate reference material.

Table 6. The amounts of reference material taking into account the production stages

Density, g/cm^3	0.3	1	1.6
Necessary amount in a unit, g	25.4	84.8	135.7
50 units, g	1272	4241	6785
200 units, g	5089	16963	27141
Bulk material for 50 units, kg	4		21
Bulk material for 200 units, kg	16		82

Among the samples detailed in Table 7, the IER2014R005 sample is sufficient for the purpose. The radiocaesium concentration is 73.4(3.2)Bq/kg which is perfect target value, because of the limit of consumption in Japan for general food is 100Bq/kg, for infant food 50Bq/kg. Taking into account the recommendation of EURACHEM/CITAC Guide (2007) [9] about "Use of uncertainty information in compliance assessment" the value 73.4(6.4) Bq/kg is clearly distinguishable from both of the limits. The isotope ratios of $^{134}\text{Cs}/^{137}\text{Cs}$ were matching the decay corrected value of the release, values varied between uncertainty range i.e. between 0.36 and 0.43 with individual uncertainties less than 10%.

Table 7: Results of the analysis of the bamboo leaves collected in 2014

sample identification	date	latitude	longitude	dose rate, nSv/h	Cs-134		Cs-137	
					a, Bq/kg	u, Bq/kg	a, Bq/kg	u, Bq/kg
IER2014R001	2014.01.14	37.679	140.4469	810	1387	12	3657	19
IER2014R004-01	2014.02.07	37.6979	140.387	262	48.8	2.5	131	4
IER2014R005	2014.02.07	37.7139	140.370	196	22.0	1.9	51.4	2.6
IER2014R009	2014.03.08	34.469	135.806	63	< 1.3		< 1.4	
IER2014R013	2014.03.11	37.255	141.013	425	7.65	0.13	19.5	0.19
IER2014R014	2014.03.12	37.683	140.452	535	21.3	1.9	56.4	2.9
IER2014R019	2014.03.17	37.808	140.508	593	102	6	282	9
IER2014R021	2014.03.19	37.3848	140.9438	1720	166	10	451	17
IER2014R024-01	2014.03.19	37.4258	141.0178	10550	1463	36	4015	59

This reference material could be used for regular checking of the performance of the laboratory. The recommended interval between the quality checking measurements is one month. For establishing the levels using of the results of 15 measurements are recommended. On the control chart around the target value 2 times of the standard deviation is necessary to use as warning level and 3 times of the standard deviation should be used as action level for the actual measuring procedure and for actual measuring system. The uncertainty of the reference material has to be also taken into account according to the uncertainty propagation law. When results are within the warning limits the method and the measurement system works reliable.

The way of the use of reference material is strongly method dependent. One reference material with several characterised target values might be used in some different ways, like in case of gamma spectrometry when density and geometry are the important properties above the target values. The determination of alpha and beta emitting isotopes requires chemical separation which is performed usually with groups of samples of same type of matrix. It is worth to put a reference material in every group of same type of samples, e.g. according to the capacity of the laboratory usually 4-20 samples are treated in same batch plus a reference material with totally same steps of the procedure. These results can be used than on the control chart of the method for proofing the reliability and a well maintained, proper use of the method.

There are many kinds of dry and instant food are available in shops of different chains of grocery stores, like the enormous variety of cup noodles, instant ramen, instant lunch, miso cups, instant yakisoba noodles; amazing that even sushi and tempura can be enjoyed as instant food. There are many kinds of dried seafood available; a lot of sauces are sold also in instant forms and so on. The density of dry bamboo leaves match with the density of these products, therefore the direct comparison is also possible.

There was no more time for finding a candidate material for the other two densities; this task is kept hopefully in the plan for near future.

Several sand samples were collected from the beaches of the seaside as the Figure 11 shows. This sample type is not sufficient for the reference material due to their low radioactivity as indicated in the Table 8. The material itself was also not a good choice at least from this region because of the big washing out of the ocean.

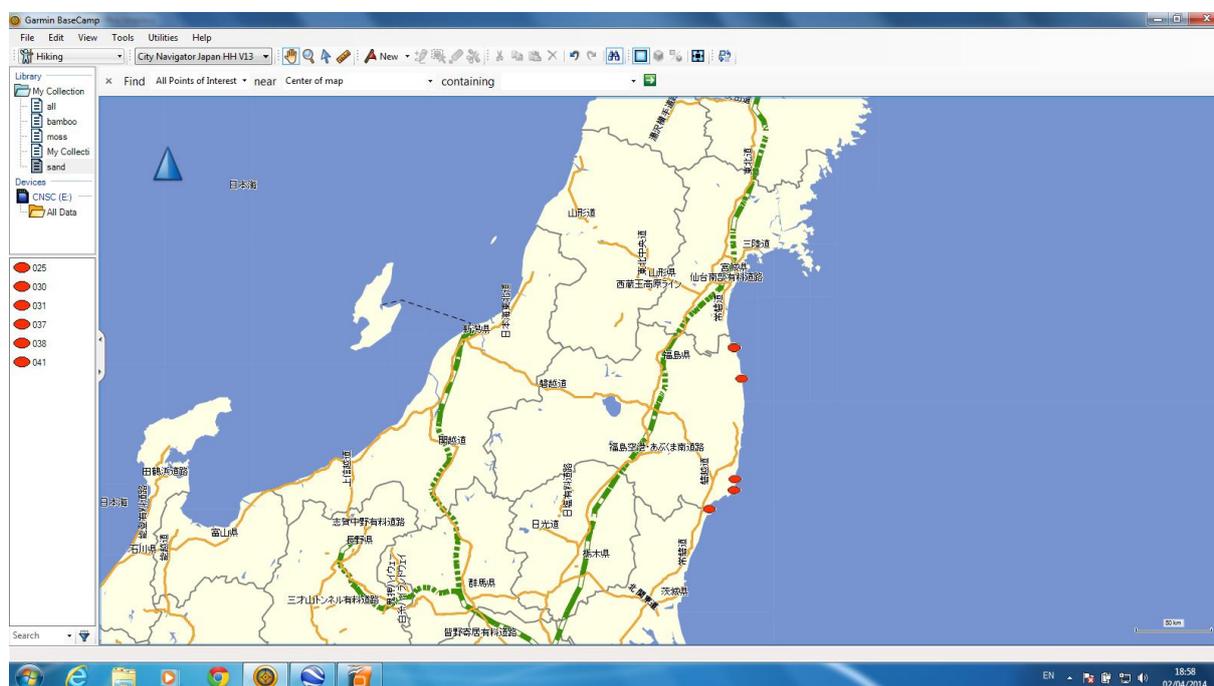


Figure 11: Sampling points of sand

Table 8: Radiocaesium content of the sand collected in 2014

sample identification	date	latitude	longitude	dose rate, nSv/h	Cs-134		Cs-137	
					a, Bq/kg	u, Bq/kg	a, Bq/kg	u, Bq/kg
IER2014R006	2014.02.24	37.042	140.974	47	< 1.08		< 1.15	
IER2014R011-01	2014.03.11	36.864	140.789	121	3.55	0.47	9.75	0.77
IER2014R012-01	2014.03.11	36.976	140.966	82	3.37	0.18	9.35	0.28
IER2014R015-01	2014.03.14	37.828	140.968	70	30.1	1.1	78.2	2.1
IER2014R016	2014.03.14	37.821	140.968	52	28.0	0.8	72.6	2.0
IER2014R018-01	2014.03.14	37.639	141.022	61	6.32	0.21	17.50	0.33

3.5 Strategy for the Assessment of the Actual Situation of Radioactivity in Japan

Complexity and fineness of pattern are characteristic of Japan's terrain. In Fukushima Prefecture Archean rock and Quaternary sediments are the main formations, but even within small areas the crust materials, their structures and the resulting landforms are often of the greatest diversity. 75% of the country is rugged hill and mountain land whose average slopes exceeds 15° which unfit for normal cultivation. The climates of Japan are a mixture of continental and marine where the continental is the dominant. Thorough the winter month's

winds in Japan are prevailingly from northwest, having originated over the cold land mass of eastern Siberia, while in summer they are from southeast, having originated over tropical seas. They are not continuous air streams, for there are frequent interruptions in the seaward flow of polar-continental air in winter and landward flow of tropical-maritime air in summer. Conclusion is that in all seasons winds blow from the several points of the compass. These climatic features result colder winter and hotter and more humid summer than are normal for Japan's latitude. There are two ocean currents a cold one from the north and a warm one from the south, but they have just slight influence upon Japan's climate. Japan is a humid land no section of which suffers from a yearly or seasonal deficiency of precipitation.

Collecting the soil samples has limited possibility in Japan because of the landcover and geographical landscape when the mapping techniques are considered for the whole country. Airborne surveys are conducted usually when the radioactivity levels are high and better not to go to the region by car – the capability of the techniques fit to this the grid is about 10km by 10km and the activity concentration is

There are some possibilities for the mapping in a detailed grid of the whole country:

1. In situ measurements with portable HPGe detectors
2. Aerosol sampling even with portable units and installed measuring stations - the second option is the proper solution around the nuclear facilities having automatic communication and warning system, these units usually belong to the so called early warning systems.
3. Collecting the bioindicators which are available in the whole country in several categories, like
 - a. Integral indicator for terrestrial environment – e.g. moss
 - b. Differential indicator for terrestrial environment – plants with big leaves e.g. any kind of leafy vegetables produces in open air conditions
 - c. Integral indicator for aquatic environment
 - d. Differential indicator for aquatic environment.

In situ measurements on the sites were performed with Ortec Micro Trans-Spec portable detector having 13% relative efficiency, the resolution at 1.33MeV is 1.92keV and the rise time is 3.4microsecundum. In most of the bamboo forest collecting soil core samples are impossible because of the very dense root-nets of bamboo, so the only solution is the in situ measurement by HPGe detectors. The situation is almost the same on the high slopes – but even the in situ data has to be treated very carefully because of the applied models usually presume an undisturbed flat area in approximately 40m circle.

Table 9: Results of the in-situ measurements performed at the beginning of 2014 in Fukushima Prefecture

file name (date,time)	Closest object	dose rate microSv/h	Cs-134		Cs-137		K-40	
			a, Bq/kg	unc, Bq/kg	a, Bq/kg	unc, Bq/kg	a, Bq/kg	unc, Bq/kg
201403191436	Otozawa Post Office	10.6	7080	1452	18540	3561	108	30
201403191340	Okuma	23.4	15284	3165	39930	7680	186	30
201403191255	Sakashita Dam	2.67	1828	396	4702	951	115	10
201403191149	Kashira Mori	1.12	292	63	761	153	229	16
201402071042	Kitayakeno	0.21	117	24.9	290	60	72.1	6.1
201402071128	Arai	0.19	56	12	145	30	140	20

201403121605	Kanyagawa	0.53	204	45	534	108	145	10
201403121645	Atago	0.81	272	60	698	141	74.6	6.3

In 2014 a pilot study was conducted for the collection of moss samples – fortunately along the big moss could be collected everywhere, the sampling points are indicated on the Figure 12, altogether 16 moss samples were collected than after drying grinded and sieved. The measuring geometry was the same as usually used in IER, so called U8 plastic container. The measurements were performed with the help of experts of Fukushima University by a measuring system equipped with auto-sampler, p-type coaxial HPGe detector with 40% relative efficiency.



Figure 12: Sampling points of the pilot study for finding proper bioindicator available in the whole country – 16 samples were collected, prepared and measured by HPGe detector

Table 10: Radiocaesium content of the moss sample collected along the country

sample identification	date	latitude	longitude	settlement	dose rate, nSv/h	Cs-134		Cs-137	
						a, Bq/kg	u, Bq/kg	a, Bq/kg	u, Bq/kg
IER2014R002-01	2014.02.02	37.661	140.495	Matsukawa	1070	3921	31	10425	50
IER2014R003	2014.01.29	37.682	140.459	Kanayagawa	760	7387	268	19320	429
IER2014R007	2014.03.02	34.836	134.693	Himeji	85	< 1.08		< 1.28	

sample identification	date	latitude	longitude	settlement	dose rate, nSv/h	Cs-134		Cs-137	
						a, Bq/kg	u, Bq/kg	a, Bq/kg	u, Bq/kg
IER2014R008	2014.03.08	34.465	135.8045	Asuka	80	< 1.08		1.30	0.34
IER2014R010	2014.03.08	34.771	136.129	Iga Uenosshi	70	< 1.08		< 1.28	
IER2014020	2014.03.19	37.383	140.941	Sakasata damn	2 670	76 525	394	209 100	647
IER2014022	2014.03.19	37.381	140.960	Kashira Mori	3 240	153 869	508	418 700	835
IER2014023	2014.03.19	37.415	140.980	Okuma	23 400	177 190	1 676	494 000	2 790
IER2014024-02	2014.03.19	37.425	141.017	Okuma	10 550	250 228	1 273	685 300	2 093
IER2014025	2014.03.21	34.401	132.460	Hiroshima	95	< 1.08		13.7	1.6
IER2014026	2014.03.22	35.013	135.75	Kyoto	75	< 1.08		18.0	2.1
IER2014027	2014.03.30	43.052	141.494	Sapporo	45	< 1.08		2.8	0.5
IER2014028	2014.03.31	39.704	141.138	Morioka	55	< 1.08		6.1	1.0
IER2014029	2014.03.31	39.001	141.101	Hiraizumi	100	< 1.08		13.9	1.7
IER2014030	2014.04.03	35.696	139.7516	Inage	70	< 1.08		9.7	1.3
IER2014031	2014.04.05	31.592	130.630	Sakurajima	56	< 1.08		< 1.28	

3.6 Suggestions for the Countrywide Monitoring in Japan

In normal circumstances the goal of the monitoring is assessing the dose for the public and maintaining the long term history of the concentration levels of the radionuclides and some other investigation for the better knowledge of the local circumstances. As it was already mentioned in the case of Hungarian countrywide monitoring, there is a need of sitting down and consider lot of governing factors in the life of the Japanese society, production rate, import of foodstuffs, consumption rate and many other parameters has to be considered. Monitoring program has to cover the agricultural production and the consumption needs of the country. There are some general issues which are not mentioned again, but the goal is to highlight the specificity, particularity, speciality of the country.

The size and the differences inside the country suggest having the monitoring program elaborated prefecture by prefecture. Japan has 47 prefectures, which definitely give good database for the statistical evaluation. Elaborating and executing the monitoring program is considerable on the region level as well, there is a practice of ordering prefectures based on their geographic regions is traditional – this solution might need less resource than by prefectures.

Table 11: Traditional geographic regions in Japan

Hokkaidō	Tōhoku	Kantō	Chūbu	Kansai	Chūgoku	Shikoku	Kyūshū
1. Hokkaidō	2. Aomori	8. Ibaraki	15. Niigata	24. Mie	31. Tottori	36. Tokushima	40. Fukuoka
	3. Iwate	9. Tochigi	16. Toyama	25. Shiga	32. Shimane	37. Kagawa	41. Saga
	4. Miyagi	10. Gunma	17. Ishikawa	26. Kyōto	33. Okayama	38. Ehime	42. Nagasaki
	5. Akita	11. Saitama	18. Fukui	27. Ōsaka	34. Hiroshima	39. Kōchi	43. Kumamoto
	6. Yamagata	12. Chiba	19. Yamanashi	28. Hyōgo	35. Yamaguchi		44. Ōita
	7. Fukushima	13. Tōkyō	20. Nagano	29. Nara			45. Miyazaki
		14. Kanagawa	21. Gifu	30. Wakayama			46. Kagoshima
			22. Shizuoka				47. Okinawa
			23. Aichi				

There are huge differences between the prefectures in size 1,862 (Kagawa) - 83453km² (Hokkaido) and in population 584,982 (Totton) – 12,059,257 (Tokyo) as well, therefore the monitoring has to contain guides for “producing” prefectures and for “consuming” prefectures and big attention has to be paid on the import control as well. The producing prefectures has to have same type of list of products as it is shown on the Figure 6 with information of passive, sowing, growing harvesting periods and they have to take into account their production. For the monitoring in consuming prefectures the main governing factor is the consumption statistics, preferably locally studied.

One of the most important statistics what has to be taken into account is the food consumption. In the Table 12 there is a part of FAO - WHO classification establishing 12 Classes on the world according to food consumption habits, where Japan belongs to Cluster L; while Hungary to cluster E; Canada, USA, Australia to Cluster M. Of course it is better to take into account the local consumption data and form the monitoring according to those data. As a first step in definition of the important foodstuff to be monitored this analysis might be useful on a general country level.

Monitoring of the total diet is not recommended, because the contamination is not supposed to evenly distributed in all kinds of foodstuffs, the source of the highly impacting food type could not be found and in such a huge country having different climatic areas and different geographical regions the diet differs a lot let us say even prefecture by prefecture.

From the point of view of the prevention and even emergency preparedness the monitoring of the raw material is preferred.

Table 12. FAO-WHO classification regarding the food consumption

GEMS	Cluster E	Cluster L	Cluster M
CEREALS	365.2	567.7	409.9
ROOTS AND TUBERS	277.4	103.6	175.7
PULSES	49.4	59.5	140.1
SUGARS AND HONEY	120.4	54.0	150.7
NUTS AND OILSEEDS	69.2	98.7	36.6
VEGETABLE OILS AND FATS	34.8	27.7	35.1
STIMULANTS	22.3	9.9	19.0
SPICES	1.8	0.6	1.7
VEGETABLES	248.6	280.5	310.5
FISH AND SEAFOOD	30.4	71.1	24.8

GEMS	Cluster E	Cluster L	Cluster M
EGGS	37.8	35.2	57.4
FRUITS	348.2	244.1	345.2
MILK AND MILK PRODUCTS	179.6	57.0	287.9
MEAT AND OFFALS	163.5	96.4	279.3
ANIMAL OILS AND FATS	15.7	4.7	24.3
BEVERAGES	312.5	85.9	298.6
TOTAL DIET IN GRAMS PER PERSON PER DAY	1929.5	1683.0	2263.1

There is a rather big difference in the consumed total amount of the food and the Japanese highest life expectancy in the world originates from their traditional food culture for sure.

If the values are normalised by the total consumption the ratios give the importance of the different foodstuffs could be seen well, on the list below only those are mentioned which has higher consumption rate than 5% recommended by IAEA [10]:

1. cereals, which is mainly rice in Japan – 34%
2. vegetables – 17%
3. fruits – 15%
4. roots and tubers – 6%
5. nuts and oil seeds – 6%
6. meet and offals – 6%
7. beverages – 5%, however the consumption rate is high not worth to deal with them, because they are processed, some of them even distilled, but if there is a high public interest the raw material has to be checked. Apriori the processing factor has to be determined in advance for different radionuclides for calculating the concentrations in consumed products.

In Japan rice has benn the main grain from early times with addition of fish and shellfish as animal protein. However fish and seafood represents only 4%, this category could not be left out from the monitoring program from several reasons, like

- some of the fishes are behaving as bioindicators for seawater and accumulate a lot from the radionuclides as well, so their proportion of the ingestion rate is expected to be higher than the proportion of the consumption,
- Japanese food uses seaweeds extensively, they have even 7 types of them identified by name and algae is almost in all prepared food in any kind of form; Polysaccharides in seaweed may be metabolized in humans through the action of bacterial gut enzymes. Such enzymes are frequently produced in Japanese population due to their consumption of seaweeds but rarer in North-American population. Since algae are rich in vitamins and minerals (up to 5% of the wet mass), almost all the deficiencies are over run by use of algae,
- Moreover there is a strong tradition behind the consumption of this category; even some people think that the artistic and cultural development of Japanese people is because of the use the seaweeds as regular, common food.

Next category – pulses – is also important from the point of view of the monitoring of the raw material: they are either eaten as dry grain or are boiled and mixed with sugar. Only for soybean do the Japanese have some kind of traditional cooking. The Japanese have many kinds of soybean processed foods including tofu, natto, miso and soy sauce. Due to the

decrease in production of soybeans in Japan and increase in their price, imports from China are preferred and American types are not suitable for natto production. This is one of the foodstuffs whose import has to be controlled well. Japan has many kinds of soy sauce but mainly two types: wheat is mixed with soybean (85% of the production) and another a little more alcoholic drink made from fermented rice with the firstly mentioned soy sauce added.

The importance of the milk product has strong geographical dependence – because of the preservation difficulties it has higher rate in nutrition in northern part of the country, where the main production is.

Tea also recommended to keep on the monitoring list because of the traditions are living around it.

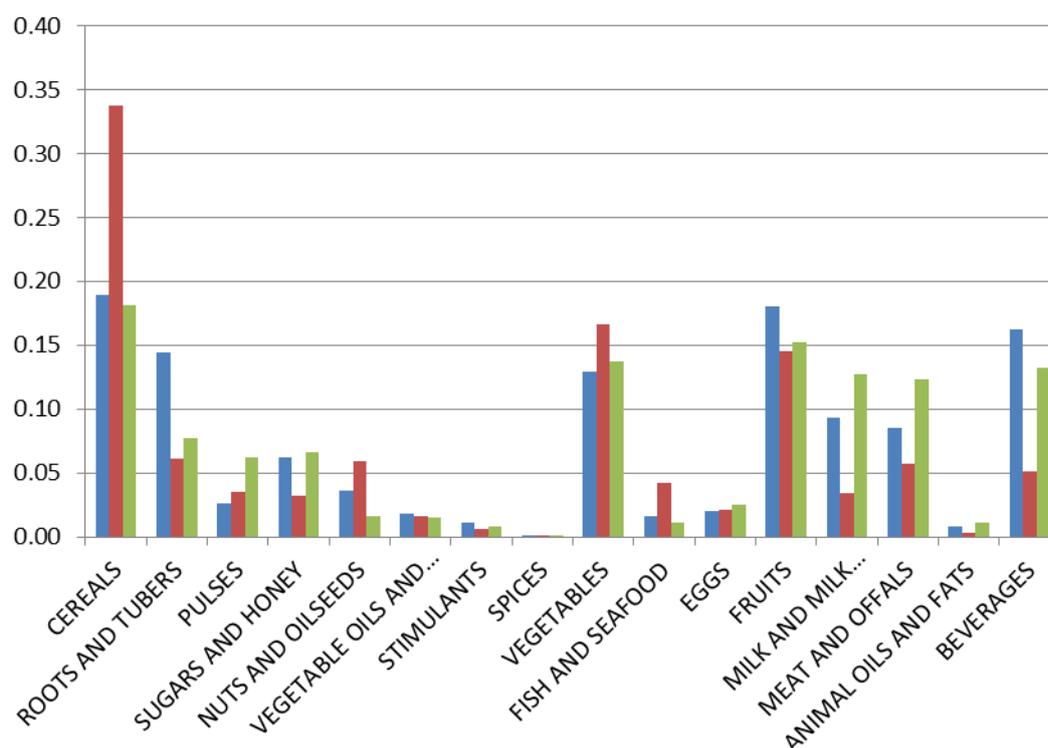


Figure 13: Relative consumption – values are normalised by the total consumed amount make visible the importance of the specific foodstuffs (blue - Cluster E, dark red – Cluster L, green – Cluster M)

In every country it is worth to think over which are the so called vulnerable products, some of them generally occurring of course and the others has some specificity:

- mushroom picked up in the forest or any kind of undisturbed area
- leafy vegetables collected as above – different kinds of fern
- wild animals, wild bores, bears, dears, rabbit
- seaweed (extremely high concentration capacity – enrichment process for minerals, special interest is from radiation point of view of I, Cs, Sr enrichment capability)
- European Union got enormous amount of the measurement data from the Japanese government, from the report of December 2013 about foodstuffs, some extremities might be pick up regarding the consumption rate (Wild Mushrooms, Ostrich fern, Aralia elata (shoot), Aralia cordata, Japanese royal fern, Pteridium aquilinum, Japanese pepper, Elatostema umbellatum var. majus (Uwabamisou), Giant butterbur, Bamboo shoot (nemagaritake), Shiitake grown on raw log

For establishing the long term series of the data the suggestion is to monitor in each prefecture:

- Rice
- Leafy vegetable, like spinach
- Root vegetable
- Seaweed
- Soybean

For having well coverage in terms of spatial distribution, same sample across the whole country has to be collected:

- Moss – integral indicator found everywhere in the country,
- Bamboo – fresh leaves could be treated as differential indicator, bamboo spot forests in different sizes are found in the country like many-many small islands.

The Table 13 contains the amounts what has to be collected for measuring the samples after some sample preparation steps not longer than a day in sufficient relative uncertainty of less than 10%. Sample preparation steps are depending on the sample type from a simple grinding to ashing, but is the usual U-8 container applied the fresh materials having dry content about 10% has to be ashed prior the one-day measurement by HPGe detectors.

Above the samples mentioned in the table installation a large volume aerosol filter with about 250m³/h airflow and big filter surface (larger than 0.2m²) would be desirable - measurement by HpGe and screening: gross-alpha, gross-beta (if necessary nuclide selective methods has to be applied for alpha and beta emitting nuclides).

Table 13: Sample amounts of foodstuffs and feedstuffs necessary to collect for measuring the actual level of anthropogenic radionuclides in Japan – 50 samples in a year in normal circumstances from each prefecture

Sample type	Sample amount in fresh weight, kg	Sample in each prefecture for a year	Remarks
Moss	2	3	Collection from flat place, like big stone, roof of the house – bigger than 1m ² continuous surface covered by moss is ideal, collection from the same area preferably
Bamboo leaves	4	3	Collection of the fresh leaves from the same place regularly
Soil	2	6	0.5m long core samples are preferred, but occasionally the upper 2cm might be enough – soil collected together with a plant is desirable for establishing the local transfer factor
Sediment	4	6	0.5m long core samples are preferred either from fresh water formation or from sea – the place of interest might modify the method of sampling, in this case the same method has to be applied on the particular spot
Water (freshwater and/or seawater)	100	6	With sufficient apparatus – measurement of dissolved part and the particulate is necessary

Sample type	Sample amount in fresh weight, kg	Sample in each prefecture for a year	Remarks
Mushroom	4	3	Preferably edible mushroom from one kind or at least from same family – grown outside The amount might seem enormous, the authority has to decide what level of confidence is desirable
Rice	1	6	Local products with well definable location of harvesting
Foodstuffs (fruits, vegetables)	4	3	Local products with well definable location of harvesting
Fish	3	3	Either freshwater or seawater fish types according to the most commonly consumed on the interested area
Seaweed	2	3	Might be cultivated as well with well-defined position of sampling
Nuts, oils seeds, dried foodstuffs, feedstuffs, mixtures, additives	1	1	Preferably local products, in case of additives at least the country of origin should be determined
Pulses	1	3	Soya bean preferably from everywhere or at least from the production site of soya product (in case of controlling the products, the processing factor has to be taken into account) – others according to the production area
Samples of animal origin with high water content either foodstuffs or any other meat	3	2	Local products with well definable location of living or kept for domestic use
Samples of animal origin with low water content either foodstuffs or any other (bones, claw, horn, antler)	1	1	Local products with well definable location of living or kept for domestic use
Fresh feedstuffs, pasture...	3	3	From 1 m ² or bigger area cut in 2 cm height, preferably without soil
Milk	5	6	Preferably from a local farm sampled together with feedstuff of animal
Wild animals	2	1	Muscle or any other edible part
Controlling import foodstuffs and feedstuffs	1-5	20	The amounts should be the same as from the local production for achieving the comparability of the measurement data

The whole analytical procedure has to be harmonised and controlled by external and internal quality measures. Harmonisation starts with the sampling through the sample preparation steps and ends with the evaluation of the data. Data of the different organisations and institutions and networks has to be comparable if they form a single database on the country level. Achieving this simply requirement is a huge, never ending work.

Methods for the determination of radionuclides: Most of the samples are processed by ashing at 450 °C, however, soil samples are air-dried, moss samples are dried at 105 °C.

1. Screening methods

- a. Gross-beta measurement, nowadays these simple counters are gradually replaced with low-background alpha-beta counters coupled to high capacity sample changer.
- b. Gross-alpha measurement: The ashed food and plant samples are measured.
- c. Gamma spectrometry by scintillation detectors are kept in routine work for checking fresh samples and also measuring ashed samples for the first quick information. For emergency situation these detectors are calibrated for measuring larger amounts of samples in fresh weight, mainly food and feed samples in 400 cm³ Marinelli beakers. However, this method is able to give activity concentration of gamma-emitting isotopes, it is considered as a screening method because of the way it is used.

2. Nuclide selective methods

- a. Determination of ⁹⁰Sr: After usual sample preparation (drying and ashing) The activity concentration might be determined after ⁹⁰Y in-growth by low-background alpha-beta counter or ICP-MS or LSC without waiting for equilibrium
- b. Gamma spectrometry by HPGe detectors in low-background shielding. Main features of the systems: coaxial detectors both n-type with the resolution of 1.8–2.2 keV, an efficiency of 30%, and able to work from 20 keV. Certified reference materials (CRM further on) has to be available with same density and same geometry as the measured samples otherwise density correction has to be applied.
- c. Alpha spectrometry by PIPS detectors: This technique together with the analytical chemistry background has to be available at least region level. The usual collecting time of alpha-spectrum is one week, it is lengthy but necessary to achieve the required sensitivity for the artificial isotopes.
- d. Liquid scintillation technique (LSC): purely beta emitting isotopes are determined, like ³H, ¹⁴C, ⁹⁰Sr...

4.0 Conclusions

In general, monitoring and sampling for radionuclides in the environment is not different from other attributes of environmental media, nor from other types of survey sampling, but never forget about the specificity of the radioactive isotopes i.e. the half-life of them might have serious effect on the sampling and therefore the monitoring or specialities occurring during the discharges also important.

Typical behaviour of the radioisotopes in the environment has to be taken into account either during the planning. Executing of sampling program and of course it will have effect on the sample preparation and the measurements. The following isotopes has the main interest because of their properties and the effect on the population and having radioecological impact: ¹³¹I, ¹³⁴Cs, ¹³⁷Cs, ⁹⁰Sr, Pu isotopes, U isotopes, ²⁴¹Am, rarely ¹⁴C and ³H as well.

These case studies have to be treated as examples and it is necessary to emphasize that other countries in another region have different needs and requirements coming from the climate, geographical circumstances, economic and social possibilities. This will require taking into account other aspects to elaborate a sufficient program.

Using the geographical information system (GIS) as well as GPS is essential in the radioecology. The set of computer tools gives possibility for collecting information, storing and retrieving them efficiently, transforming and displaying spatial data i.e. measurement results placed on the right position of the map.

There might be some additional political, social or economical reasons for executing sampling in some locations, irrespective of what might be optimal for a model based network design.

5.0 Acknowledgements

This work was performed in 2014 at Institute of Environmental Radioactivity at Fukushima University. The corresponding author is very grateful for the management of the IER, management of the Fukushima University and management of the Central Agricultural Office for the great possibility of working three months in IER and was pleased to enjoy the warmness, helpfulness of all the staff members there.

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