



This project has received funding from the Euratom research and training programme 2014-2018 under grant agreement No 662287.



EJP-CONCERT

European Joint Programme for the Integration of Radiation Protection Research

H2020 – 662287

Guidance to reduce sampling uncertainty (D9.60)

Application to radiological monitoring

Lead Author: Juan Carlos Mora & Almudena Real (CIEMAT)

With contributions from: Pedram Masoudi, Mathieu Le Coz, Charlotte Cazala, Mathilde Zebracki, Arnaud Mangeret, Marie Simon-Cornu (IRSN); Jordi Vives i Batlle (SCK-CEN); Mark Dowdall (DSA); Danyl Pérez-Sánchez (CIEMAT); Antti Kallio (STUK); Martin Steiner (BfS)

Reviewer(s): CONCERT Coordination team

Work package / Task	WP 9	T9.1 (TERRITORIES)	SST 9.3.1.1
Deliverable nature:	Report		
Dissemination level: (Confidentiality)	Public		
Contractual delivery date:	M50		
Actual delivery date:	M50		
Version:	1.0		
Total number of pages:	39		
Keywords:	Sampling uncertainty, sampling strategies, radiological monitoring, guidance		
Approved by the coordinator:	M50		
Submitted to EC by the coordinator:	M50		

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Abstract

This report presents part of the work undertaken in CONCERT sub-subtask 9.3.1.1 (i.e. TERRITORIES task 1.1), which addresses radiological characterisation of long-term contaminated territories. This radiological characterisation can be based on multiple monitoring resources and therefore will depend on the methods used, the spatial and temporal integration scales applied, the actors performing the measurements and the objectives of the monitoring campaign. Some general aspects about monitoring have already been covered in a previous CONCERT-TERRITORIES deliverable (CONCERT-TERRITORIES D9.74, 2018), which included some terminology discussions about the words “sampling” and “monitoring”, and provided general recommendations about the sampling of a material from an environment, its transport to the laboratory, preservation and traceability. Therefore such aspects will not be covered in the present document. The deliverable D9.60 addresses sampling strategies, sampling uncertainties, and how to reduce them to better assess spatial and temporal variability. In this document, the word “sampling” is used with its statistical definition, i.e. the selection of a set of units or elements from a larger population, typically to be observed to make inferences regarding that population and “monitoring” is used in a broader sense, including all methods, actors and objectives mentioned above. Several documents have addressed the description of uncertainties associated with the radiological characterization of territories, including measurement and sampling uncertainties, as well as the numerical methods available to optimize the number of samples needed for a good characterization in order to have a good understanding of variability. The present document D9.60 intends to develop a summary of the most used techniques for characterizing sampling uncertainty, providing technical guidance on their practical implementation, and some recommendations on reduction of sampling uncertainty.

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1 INTRODUCTION: TERMINOLOGY AND SCOPE OF THE DOCUMENT

CONCERT sub-subtask 9.3.1.1 (i.e. TERRITORIES task 1.1) addresses the radiological characterisation of long-term contaminated territories. This radiological characterisation can be based on multiple monitoring resources, differing by the methods and hence the spatial and temporal integration scales (from traditional field sampling followed by laboratory measurements to mobile *in situ* monitoring), by the actors performing the measurements (e.g. environmental monitoring professionals, researchers, lay people through monitoring crowdsourcing), and by the objectives (e.g. for regulatory purposes, to calibrate predictive models). Case studies of the TERRITORIES project illustrate this diversity, as detailed in another deliverable of the project introducing the TERRITORIES Library Database (CONCERT-TERRITORIES D9.59, 2019), and in publications and communications (Crouail et al., 2018, 2019; Masoudi et al., 2018, 2019, submitted; Zebracki et al., 2019).

Some general aspects about monitoring have already been covered by a previous TERRITORIES deliverable (CONCERT-TERRITORIES D9.74, 2018), resulting from a workshop held in Madrid in June 2018. Workshop discussions, tracked in the deliverable, had included some terminology discussions about the words “sampling” and “monitoring”. “Sampling” appeared to be more related to the physical taking of some material portion from the system under study or environment (e.g. soil, air, water) to be analysed or tested in the laboratory (not *in situ*), and/or appeared to be more related to research activities (to find data for modelling, spatial distribution), whereas “monitoring” appeared to be more related to operational/routine surveillance in a time span (to demonstrate compliance with regulation and for assessment purposes), and/or *in situ* measurements (installing some instruments for surveying the changes over time). The deliverable D9.74 also provided general recommendations, including some technical aspects about the sampling (physical taking) of a material from an environment (soil, air, water), its transport to the laboratory, preservation and traceability. The reader is invited to refer to this document D9.74 as such aspects will not be covered by the present document, D9.60.

Indeed, the present D9.60 document addresses sampling strategies, sampling uncertainties, and how to reduce them to better assess spatial and temporal variability. In this document, the word “sampling” is used with its statistical definition, i.e. the selection of a set of units or elements from a larger population, typically to be observed to make inferences regarding that population (according to US EPA, 2002) and “monitoring” is used in a broader sense, including all methods, actors and objectives mentioned above.

Obviously this is not the first document on the characterization of uncertainties associated with the radiological characterization of territories, and even the International Committee on Radiation Units (ICRU) and the International Atomic Energy Agency (IAEA) have published technical documents or chapters on this subject (IAEA, 2004; ICRU, 2006). A lot has been written on measurement uncertainty (GUM, 2008) and many laboratories implement adequate protocols on the issue. However, natural variabilities must also be adequately characterized and the uncertainty resulting from sampling variable elements should be recognized and quantified (Eurachem, 2007). A good understanding of variability is also important, and many approaches exist for the problem, providing even numerical methods to optimize the number of samples needed for a good characterization (MARSSIM, 2000; US EPA, 2002). The deliverable D9.60 intends to develop a summary of the most used techniques for characterizing and quantifying sampling uncertainty, to provide technical guidance on their practical implementation, as well as some recommendations on reducing sampling uncertainty.

Chapter 2 describes the sampling uncertainties, including the two early theories, statistics and probability theory, which address this challenge. Chapter 3 explains basic concepts and principles of sampling, summarising them in a sequential five-step approach. Chapter 4 includes a brief description of the different methods available for sampling design and quantifying the uncertainty of sampling. Chapter 5 is dedicated to 3 case studies from the TERRITORIES Library: a Belgian NORM forest, the Fukushima area and a Polish Lake contaminated with NORM. Chapter 6 presents the conclusions and guidelines based on the work done for the deliverable D9.60.

2 SAMPLING UNCERTAINTIES

Spatial, temporal and inherent variabilities sum up to total observed variations between individual, single samples. Due to the variability, every individual sample of the whole population will not be identical. Therefore, a proper sampling strategy is needed to achieve accurate values of the desired quantity (e.g. ambient dose rate; activity concentration of radionuclides).

In order to separate sampling uncertainty from measurement uncertainties, it is assumed that every step of collecting and measuring the given quantity is accurately executed.

Two early theories, which address the challenge of sampling uncertainty, are statistics and probability theory. **Statistical theory** proposes statistical parameters, e.g. variance, skewness and kurtosis, for studying the variability of the real value by samples. But how many samples should be acquired to have a certain confidence level of the statistical parameters? **Probability theory** proposes a solution to this question by considering a specific density function for the population under study. So the minimum number of samples needed in order to achieve a certain confidence level could be determined. At the same time another question arises: Are the samples representative of the population? This question is addressed in each domain differently, e.g. random sampling in regular mesh grids was proposed in the geochemical studies related to mineral explorations; or in sociology the samples should keep the proportions of social classes in order to prevent biased results.

3 BASIC CONCEPTS AND PRINCIPLES OF SAMPLING

The basic concepts and principles of sampling can be summarized with a sequential five-step approach:

- i) Define the objectives and questions to be answered.
- ii) Summarize the environmental context for the quantities being measured.
- iii) Identify the target population, including spatial and temporal extent.
- iv) Select an appropriate sampling design.
- v) Document the sampling design and its rationale.

Note that in some cases, the sampling strategy is inherently given, i.e. not a deliberate choice. This is for example the case with a meta-analysis of the literature (illustrated in the TERRITORIES Library by the study of the Fukushima forests in Japan), or of monitoring crowdsourcing literature (illustrated in the TERRITORIES citizen science study in Belarus (Crouail et al., 2018, 2019)). Such cases will not be further developed in the rest of the document, which will from now focus on deliberate sampling strategies.

i) Define the objective and questions to be answered

The purpose(s) of the sampling program must be clearly specified before the sampling program is designed, because different purposes require different sampling strategies, sample numbers and exigencies in order to be efficient.

Purpose(s) include:

- Characterizing a variable, e.g. ambient dose rate [$\text{Sv}\cdot\text{h}^{-1}$], or radionuclide deposition [$\text{Bq}\cdot\text{m}^{-2}$], or mass activity density (concentration) of a radionuclide in sediment, water, or biological tissue [$\text{Bq}\cdot\text{kg}^{-1}$], in a population (or area), and more specifically:
 - assessing its (arithmetic or geometric) average or the most probable value,
 - describing its range (or magnitude) of variability, or assessing its (arithmetic or geometric) standard deviation,
 - mapping its spatial variability (e.g. detecting a spatial trend, or locating hot spots, or cartography of a map),
 - describing its temporal variability (e.g. detecting a temporal trend),
 - quantifying the proportion of area above a threshold (e.g. background, a regulatory limit or a specified intervention level).
- Comparing two (or more) populations/areas for one variable.
- Correlating two (or more) variables for one population/area.

Such purposes cover various objectives, as for example assessing compliance with regulations, assessing the consequences on the human health or environmental impacts, estimating radioactive waste volume (see the CONCERT-TERRITORIES D9.74 for a more exhaustive review). Many experimental and monitoring programs have multiple objectives; in this case, the relative importance of each objective should be identified. Sampling design is a compromise between sample cost and sample value. The latter should be approximated based on the objective of the campaign and its importance (Desnoyers and Dubot, 2014a, 2014b).

ii) Summarize the environmental context for the quantities being measured

Statistical sampling requires information about the nature of the population and a description of its characteristics. The environmental context provides four general sorts of information:

- nature of the population such as the physical or biological material of interest, its spatial extent, its temporal stability and other important characteristics,
- expected behaviour and environmental properties of the radionuclide of interest in the population,
- sampling unit (i.e., individual sample or specimen), meaning the individual or defined portion of a medium selected from the population for measurement,
- expected pattern and magnitude of variability in the observations.

It is often the case that a pilot study would be necessary to provide a preliminary estimate of the variability in the population. Alternatively, one could proceed with a presumed estimate of the expected variability.

iii) Identify the target population, including spatial and temporal extent

In the frame of this document, the population of interest is expected to be an area. A practical issue is the dimension of this area which varies from a limited parcel at a local scale (cf. case study of § 5.1 for the Belgian NORM site) to a post-accidental situation (Sanada et al., 2014; Saito and Onda, 2015; Saito et al., 2019; Case-study of §5.2) at a regional or global scale.

Temporal evolution is expected to be smooth in the context of this research project, as TERRITORIES is concerned by long-term contamination, apart from countermeasures that may immediately change the radiological situation.

iv) Select an appropriate sampling design

The choice of sampling design often represents a compromise among four different goals: estimability, precision, efficiency and defensibility.

Chapter 4 of this document is dedicated to sampling design methods.

v) Document the sampling design and its rationale

A quality control and assurance protocol should be part of every sampling program, during sampling design, sample collection, handling and analysis. A quality control document for an environmental sampling program should contain at least the following information:

- clear description of the objectives of the study,
- explicit definition of the target dimension, shape and sampled population,
- required confidence level, degree of accuracy, precision and spatial resolution,
- sampling date, sampler name and other environmental specifications (e.g. temperature and humidity if necessary),
- specifications of random generator(if any) and its robustness,
- metadata: a brief description of location area, physical or temporal conditions that might be effective on the results,
- probable biasedness, and
- probable uncertainty sources.

Regarding the case studies of the TERRITORIES Library, such information can be found in another deliverable, CONCERT-TERRITORIES D9.59 (2019), describing the TERRITORIES Library database (abbreviated TLD), or for 3 case-studies in § 5.1 for the Belgian NORM site, § 5.2 for the flight lines and § 5.3 for a NORM Polish Lake.

4 SAMPLING DESIGN: METHODS TO CHARACTERISE AND QUANTIFY UNCERTAINTIES

4.1 Methods to characterise sampling uncertainty

Several methods exist to design representative sampling campaigns with an optimum number of samples. Probability-based sampling is also able to choose a set of extreme samples. See NUREG-1505 for this numerically based design of sampling (US NRC, 1998). Some accepted methods for the sampling design were described in the ICRU 75 (2006). A brief description of them is included below.

4.1.1 *Judgmental sampling*

It is usual to design a sampling subjectively, i.e. based on expert reasoning and judgment, without a strict mathematical prove. The main issue in subjective sampling design is that each expert might prefer his individual pattern which might bring different monitoring results, being difficult to prioritize different designs. In other words, it is difficult to judge the representativeness of the samples quantitatively. Other disadvantages of subjective sampling include: (i) there is no idea about the optimum number of samples, i.e. either too much or too few, (ii) possibility of missing the hot spots and small anomalies, and (iii) possibility of biased sampling.

It should be noted that subjective decision making is sometimes inevitable, e.g. in (i) totally new conditions for which enough a priori information or data are not available; (ii) the very first phases of characterization; and (iii) high activity densities of radionuclides caused by known effects, e.g. sampling near the end of a discharge pipe.

4.1.2 *Systematic sampling on grid*

Systematic sampling is probably the most commonly used method for field sampling. It is generally unbiased as long as the starting point is randomly selected and the rules to select the samples are systematically followed. This method is very simple to implement in practice. However if the initial judgment to select the number of samples or the sampling rate are not correct, important characteristics might be missed during the process.

For example, a grid is considered as being overlaid (rectangular or otherwise) in a given site, and sampling locations are on gridline intersections at a fixed distance apart in each of the two directions. The starting location is expected to be randomly selected. If in a given rectangular area with dimensions X m and Y m, a number of samples $X \times Y$ are decided to be sampled, where the total area is divided into unitary areas, and the location for the first sample is randomly selected in the first unitary corner area; then subsequent samples are located at multiples of 1 m from the first location in the two directions. So all samples' coordinates are conditioned to the first corner coordinates. In order to make the samples' coordinates independent from the first corner, sampling could be done randomly inside each grid cell. In this example, the hot spots, smaller than the sampling mesh or sampling rate, i.e. 1 m, might be missed. The value of the samples and the probability of characterizing a hot spot which is smaller than the sampling rate could be expressed by Nyquist frequency, which is one over sampling rate.

If the spatial variation is uniform in both directions (along X and along Y), the environment is called isotropic and uniform sampling is the best choice. If the spatial variability is higher in one direction, compared to the other, it is called geometrical anisotropy. Along the high-variability direction, the sampling should be denser than along the low-variability direction, i.e. perpendicular to the direction of the contaminating plume or perpendicular to the geological strata, sampling must be relatively denser, e.g. every 0.5 m along the high-variability direction and every 2 m along the low-variability direction. In this example the number of samples along the direction with high spatial variability (f_{Nyq}^h) is four times higher than the number of samples along the direction with low spatial variability (f_{Nyq}^l). In other words, the probability of characterizing a unidimensional target (which is smaller than the sampling rates) is four times higher in direction f_{Nyq}^h . The targets larger than the sampling rates would be always characterized by the sampling pattern.

$$f_{Nyq}^h = \frac{1}{0.5} \text{ and } f_{Nyq}^l = \frac{1}{2}$$

A systematic grid may also include additional sampling points at distances shorter than the grid cell size. These small-distance sampling points can provide additional information about small-scale spatial correlations.

Sampling exactly at the points of a regular grid is prone to any periodic pattern in the quantity to be investigated if the periodicities of the regular grid and the sampled quantity agree by chance. A variant, i.e. the 'unaligned grid' pattern, reduces the potential bias resulting from a periodicity.

If spatial variation is not constant in different sampling zones, it is called zonal anisotropy. It is logical to have denser sampling in the zones with higher spatial variability to be able to record the maximum possible variability. From a practical point of view, in the first phase, the less dense sampling grid could be designed according to the zones with lesser spatial variability. In the second and further phases, denser sampling could be done locally if necessary.

4.1.3 Random sampling

In random sampling strategy, samples' locations are indicated by a random generator program through a uniform probability density function. Therefore, every location or individual within the population has exactly the same probability to be sampled. The sampling is unbiased, however might not be representative of spatial variability if geometrical or zonal anisotropy exist.

In cases where some characteristics of the population or the area to be sampled are known (as the types of soils in a given area), stratified sampling is more convenient, where the number of samples to be collected in each soil type could be defined accordingly.

4.1.4 Stratified sampling

Stratified sampling is more efficient in the environments with zonal anisotropy. In this method the total population or area is subdivided according to their known characteristics. Subdivision in stratified sampling is based on geological or experimental indicators. Thereafter, a random sampling can be used inside each subdivision or subarea. In subareas where spatial variability is higher or where more

accurate characterization is required, the number of samples could be increased. In stratified sampling, a priori knowledge of the population/area is needed. Since sampling is conditioned to the subdivision, any mistake in defining subdivisions would increase the risk of biasedness.

4.1.5 Cluster sampling

Cluster sampling is applied where members of the population or characteristics in an area are found in clusters or colonies. Cluster sampling involves dividing the population into primary units. Each primary unit is then divided into secondary units. A sampling design (e.g., a simple random sample or a systematic design) is used to select a subset of the primary units. Then, all the sub-units in the chosen primary units are sampled.

Clusters of individuals are selected randomly and all individuals within each cluster are selected and measured. Another variant would involve random selection of a fraction of the individuals within a cluster.

As an example, consider sampling lichens growing on boulders scattered in a forest. It would be difficult and perhaps impossible to enumerate all the lichens from which to draw a simple random sample. A systematic sample would be a problem because there may not be any lichen at many of the grid points. A cluster sample then becomes a reasonable sampling method. The boulders are the primary units. The secondary units are the lichens. Estimation of the population mean and the standard error of the mean is relatively easy for a cluster sample in which all secondary units in a selected primary unit are collected for measurement. The data are reduced to totals for each primary unit, then analysed as appropriate for the sampling design used to select the primary units.

A variant of the cluster sample is a two-stage sample. A subset of primary units is sampled, just as for a cluster sample. However, in a two-stage design, only a subset of the secondary units on the selected primary units is sampled. Because there are two levels of random sampling in this case, the estimation of the population mean and the standard error of the mean is more complicated. Adaptive sampling is also a form of cluster sampling in which decisions are made during the survey, particularly when a cluster, such as hot spots, is detected unexpectedly.

4.1.6 Composite sampling

Compositing is a technique for reducing the variability among sample units, especially useful when (i) the cost of sample processing is high, (ii) the spatial variability of adjacent samples is not within the monitoring objectives, while it is important to know the total amount of a contaminant, or (iii) the natural randomness, i.e. uncertainty exists in the samples, so integrating the subsamples neutralizes the uncertainty and results in unbiasedness.

In composite sampling, a specified number of sub-samples are collected from a single sampling unit and combined into a single sample before analysis. The measurement result of the analysed composite sample is expected to be the same (assuming very low measurement error) as the average of the measurement results of the sub-samples, but this 'average' is obtained mechanically by mixing the sub-samples and performing a single analysis on the composite sample. For example, in geochemical sampling from river sediments, if we are only interested in spatial variability along the river (and not

across the river), three sediment sub-samples from three different distances from the river bank could be acquired and mixed to analyse a single composite sediment sample for each profile along the river.

As a temporal example, a composite sample could be prepared by integrating the sampled air filters over 12 months, if seasonal variability is not within the scope of sampling or if the samples are too much heterogeneous in short timespans.

Collecting a larger sample or combining temporal samples is a specific type of compositing, e.g. combining all the 12 monthly sampled air filters into a single measurement. If the material is heterogeneous with considerable small-scale spatial variation, the variability among samples is likely to be larger than that among composite samples.

4.1.7 Sampling location candidates based on geostatistical tools: Kriging methods

Monitoring is sometimes performed in different phases or is repeated periodically, like Fukushima airborne radiation monitoring. Therefore, sampling in later projects could be readjusted according to the results of previous sampling campaigns. Geostatistics provides some criteria to complete previous sampling campaigns conditioned to the available information.

For the monitoring objective of cartography or producing a map, e.g. contamination map or air dose rate variation, interpolation methods are used. Kriging methods are geostatistical interpolation tools which are used to interpolate (estimate) a variable on a regular grid (unobserved points), according to the sampled locations (observed points). Compared to other interpolation methods, a big advantage of kriging methods is in considering spatial correlation of the variable, zonal and geometric anisotropies through variogram analysis. A variogram also quantifies uncertainty or natural randomness of the samples (called nugget effect). In parallel to interpolating the variable, kriging calculates the interpolation (estimation) error. Kriging is known as the Best Linear Unbiased Estimator (BLUE) since it minimizes the kriging variance, i.e. the interpolation error. Kriging variance is a function of the samples configuration and spatial correlation of the variable, i.e. the variogram. Locations with high kriging variance are candidates for further sampling campaigns. In the literature, minimizing kriging variance is used as a criterion for designing sampling pattern or in suggesting new sampling locations, examples are listed in Table 1.

In some sampling problems, it is argued that minimizing only kriging variance is not the best criterion for sampling design, especially in the case of non-stationary data where spatial variation is not constant over the space. In the non-stationary situation, it is suggested to couple kriging variance with a measure of spatial variability in order to increase the sampling density in areas where spatial variability increases (Delmelle and Goovaerts, 2009). Another solution of the non-stationary problem is to replace kriging variance by uncertainty of the variable, reconstructed by conditional geostatistical simulation (Hernández and Emery, 2009).

In some sampling applications, having a precise characterization is not the final purpose, e.g. for proposing additional borehole drilling points in the Shah-Kuh Pb-Zn deposit, west central Iran, it is suggested to maximize a GET function: multiplication of average estimated block grade (G) and total ore thickness (T), divided by average kriging variance (E) (Hassanipak and Sharafodin, 2004). Maximizing the GET function means proposing boreholes drilling for the purpose of increasing reliability and decreasing interpolation error at the same time.

The GET function was used as an objective function in order to propose infill drilling locations in Kahang Cu porphyry deposit, northeast Isfahan, central Iran (Morshedy and Memarian, 2015). For proposing infill drilling locations in Choghart apatite-bearing iron oxide deposit in central Iran, a new objective function is proposed which considers kriging variance and ore value function (economic price of iron and phosphor) simultaneously (Morshedy et al., 2015).

Table 1. Examples of minimizing kriging variance for sampling point selection.

Application	Brief of proposed method or index	Reference
Infill sampling design for estimation of oil rock thickness, China	Proposing Relative Updated Kriging Variance (RUKV)	Gao et al., 1996
Pb-pollution characterization in an urban region	Spatial simulated annealing for minimizing kriging variance	van Groenigen and Stein, 1998
Texture and phosphate content on a river terrace, Thailand	Spatial simulated annealing for minimizing kriging variance	van Groenigen et al., 1999
Water table in a cover-sand area, Netherlands	Simulated annealing for minimizing spatially Averaged Universal Kriging Variance (MUKV). Covariates: relative altitude, drainage depth and drainage density	Brus and Heuvelink, 2007
Contaminated soil by heavy-metal Ni, Taiwan	Adaptive cluster sampling based on regulation threshold and kriging variance, for additional sampling. The new sampling points were wherever Ni concentration is close to the regulation threshold, while first-phase sampling density is low	Juang et al., 2008
Optimizing (reducing) gamma-radiation monitoring stations, European countries	Minimizing spatially averaged regression kriging standard deviation	Melles et al., 2008
Mapping four soil properties, central Czech Republic	Minimizing sample size while keeping kriging variance below a threshold: modelling mutual spatial dependence of the soil properties by linear model of co-regionalization (cokriging), computing spatial coverage sample by a clustering algorithm, and optimizing sampling design by simulated annealing	Vašát et al., 2010
Air quality measurement survey in urban areas, contaminant: benzene or nitrogen dioxide, Bordeaux agglomeration, France	Standard simulated annealing and interacting particles algorithm for minimizing universal kriging variance; auxiliary variable is included using a covariate model	Romary et al., 2014
Monitoring quality of cultivated land, Beijing Daxing District, China	Spatial simulated annealing for minimizing kriging variance	Tang et al., 2014
Estimating an ecological index, regional gross primary production, Babao river basin, China	Minimizing spatially averaged kriging variance by a spatial simulated annealing search algorithm	Wang et al., 2014
Airborne measurement of soil properties, hypothetical case study	Finding a trade-off between kriging variance and total distance walked to visit all points by AMOSA algorithm	Lark, 2016
Adding or deleting sampling locations for characterizing sodium concentration in groundwater, Punjab, Pakistan	Using spatial simulated annealing for minimizing mean universal kriging variance	Zahid et al., 2016
Infill directional drilling design, Dalli Cu-Au deposit, Iran	Minimizing kriging variance by particle swarm optimization algorithm	Fatehi et al., 2017
Rain-gauge location for rainfall prediction, north-east of the city of Manchester, UK	Minimizing variance of kriging with external drift by spatial simulated annealing	Wadoux et al., 2017

Considering a fictitious nuclear accident at the Borssele nuclear facility, Netherlands, it is illustrated that minimizing kriging variance is not the best cost function for radioactivity monitoring and mapping. It is explained that in a nuclear accident, classifying contaminated zones is much more important than

characterizing the exact values of contamination; therefore it is suggested to minimize the costs of wrong decisions caused by areas with false classification, especially false negatives areas, since the costs associated with false negatives are higher than those associated with false positives. Minimizing the cost function by simulated annealing suggested implementing new temporary monitoring stations in the areas where the contamination level is close to the intervention threshold (Heuvelink et al., 2010). In another case study, the permanent network of radiation monitoring was optimized in the Netherlands and Germany by minimizing the sum of kriging variance and cost of failing to detect a plume by at least two detectors within three hours after a nuclear accident (Melles et al., 2011).

4.2 Methods to quantify sampling uncertainty

Sampling is an integral part of the measurement process. There is evidence that sampling is often contributing more to the total uncertainty of measurements than the analytical part of the measurement process, and therefore the uncertainty arising from the sampling process must be evaluated. Eurachem has developed specific guidance on the procedures for estimating the resulting sampling uncertainty for chemical contaminants (Table 2), which is also applicable to radioactive pollutants (Eurachem, 2007).

Table 2. Sources of sampling uncertainty in sampling theory (Eurachem, 2007).

Source	Description
Fundamental sampling error	A result of the constitutional heterogeneity (the particles being chemically or physically different)
Grouping and segregation error	A result of the distributional heterogeneity
Long-range point selection error	Trends across space or over time
Periodic point selection error	Periodic levels across space or over time
Increment delimitation error	Identifying the correct sample to take. Considers the volume boundaries of a correct sampling device
Increment extraction error	Removing the intended sample. Considers the shape of the sampling device cutting edges
Increment and sample preparation error	<i>Contamination</i> (extraneous material in sample) <i>Losses</i> (adsorption, condensation, precipitation, etc.) <i>Alteration of chemical composition</i> (preservation) <i>Alteration of physical composition</i> (agglomeration, breaking of particles, moisture etc.) * <i>Involuntary mistakes</i> (mixed sample numbers, lack of knowledge, negligence) * <i>Deliberate faults</i> (salting of gold ores, deliberate errors in increment delimitation, forgery, etc.)
Weighting error	The result of errors in assigning weights to different parts of an unequal composite sample

*Excluded from uncertainty estimates as gross errors.

The Guide describes various methods that can be used to estimate the uncertainty of the measurement process, particularly that arising from the processes of sampling and the physical preparation of samples. The reader is invited to refer to this guidance for more details and practical examples in different application areas. The present document, D9.60, only includes a summary of the main approaches and issues to be considered to quantify sampling uncertainty.

Sampling uncertainty can be estimated using two main approaches:

- The empirical approach (also called experimental, retrospective, or top-down) uses repeated sampling and analysis, under various conditions, to quantify the effects caused by factors such as

the heterogeneity of the analyte in the sampling target and variations in the application of one or more sampling protocols. It quantifies sampling uncertainty and usually the relevance of some of its components (Table 3).

- The modelling approach (also called theoretical, predictive or bottom-up) uses a predefined model that identifies each of the component parts of the uncertainty, making estimates of each component, and sums them in order to have an overall estimate.

Table 3. Empirical methods used to estimate combined uncertainty, including sampling uncertainty (Table adapted from Eurachem, 2007).

Method #	Method description	Samplers (person)	Protocols	Component estimated			
				Sampling		Analytical	
				Precision	Bias	Precision	Bias
1	Duplicates	Single	Single	Yes	No	Yes	No ¹
2	Protocols	Single	Multiple	Between protocols		Yes	No ¹
3	Collaborative trial in sampling	Multiple	Single	Between protocols		Yes	Yes ²
4	Sampling proficiency test	Multiple	Multiple	Between protocols + between samples		Yes	Yes ²

¹Analytical bias information may be obtained by including certified reference materials in the analytical run.

²Analytical bias is partially or completely included in collaborative exercises where multiple laboratories are involved.

Guidance is given in the Eurachem report (2007) on the selection of the most appropriate approach for any application. The extra cost of estimating uncertainty is also considered in relation to the cost savings that can be made by knowing the uncertainty of measurement more reliably. However, the authors recognise that such a Guide can never be fully comprehensive, and there will often be a need for expert advice in more complex situations.

5 CASE STUDIES

5.1 A case study: Uncertainties in sampling and monitoring in a Belgian NORM contaminated forest

5.1.1 Background

Radioecological sampling and monitoring in forest ecosystems is fraught with important challenges. The forest ecosystem is a complex open environment, comprising many interactions from the soil to the vegetation to the atmosphere, and there are a large number of processes associated with the fate and transport of the radionuclides such as hydrological processes, vegetation processes, atmospheric processes and the linkage of radionuclides to water fluxes and energy fluxes within the system. The issue is further compounded by the fact that the forest vegetation (operating under the influence of time-dependent climatic variables) exerts an important effect on the water cycling within the system. The forest environment is subject to extensive spatial and temporal heterogeneities and for all the above reasons it is a notoriously difficult system to sample and monitor.

5.1.2 Main sources of uncertainty

The first challenge in forest sampling and monitoring is the identification of a suitable site within the forest environment. This means that it is necessary to carry out an initial characterisation survey to identify the portion of the forest to be investigated.

The primary source of uncertainty here is the spatial heterogeneity of the contamination. Relatively high contamination areas may be very close to areas where the levels are very low, almost down to background, with the presence of occasional hot spots possibly within a few metres. This uncertainty is addressed by carrying out a radiological gamma-ray walking or aerial surveys of the wider area. In the course of investigations of the Belgian NORM site for this project, one such a survey was carried out (Figure 1).

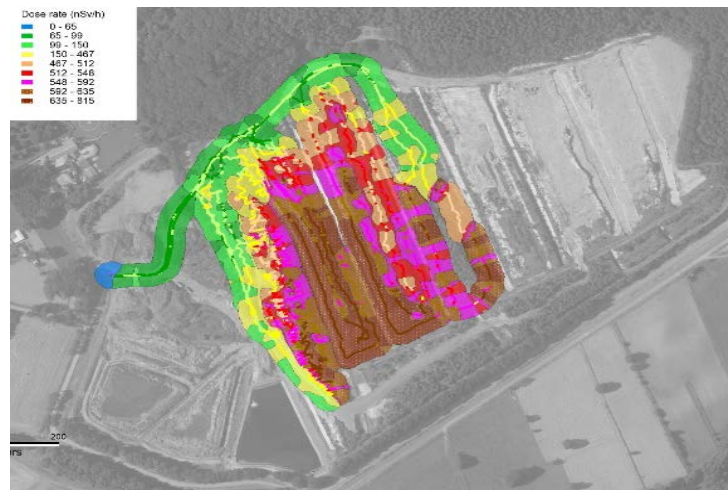


Figure 1. Uncertainty due to spatial heterogeneity in soil contamination, as evidenced by gamma-ray dose survey, in the Belgian NORM site.

Another source of sampling uncertainty is the heterogeneity of the vegetation in terms of variability of plant species, morphological and age differences between the same species of trees and health status of the trees. This uncertainty is addressed by carrying out species identification, assessing health status (height of canopy, density of branches and straightness of the tree trunk), determining the key dendrometric parameters (principally tree circumference at breast height) from which trees belonging to a typical even aged distribution can be statistically identified, and assessing the presence of other types of vegetation and understory. In the course of TERRITORIES, such a survey has already been carried out, using as a bio-indicator species the Scots pine tree (*Pinus sylvestris*) (Figure 2).

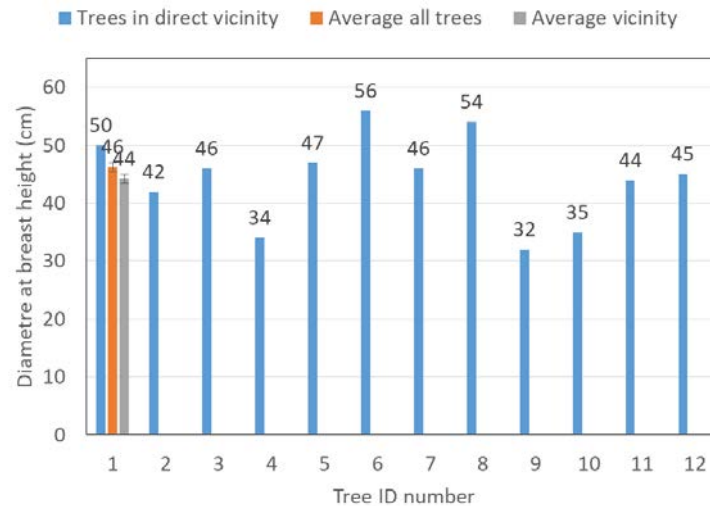


Figure 2. Dimensions of 12 Pine trees in the immediate vicinity of the Belgian NORM site monitoring station, compared with the representative average of all the trees in the wider area.

The third main source of uncertainty is the heterogeneity of the soil (Figure 3). This is addressed by performing soil characterisation: mineralogical and chemical composition, vertical layering (with the identification of the physical-chemical properties of the soil horizons using standard methodologies), and examining soil water retention in order to estimate the fractions of retained and extractable water. In terms of soil characterisation, the sludge at the Belgian NORM site is a relatively impermeable layer, covered by ~ 10 cm of black organic soil. The sludge does not look capable of fast drainage, because it is quite fine and compact and there is little air inside. Hence, the sludge acts as an impermeable layer, accumulating water in winter and slowly drying in the summer by evapotranspiration. The sludge appears visibly low in sand content (probably less than 10%) and is likely highly depleted in nutrients (e.g. phosphates) and also likely to have chemical pollution (to be determined in further analyses). The combination of anoxic soil, bad nutrition, poor nutrient balance and chemical pollution may be the cause of the poor health observed in the trees.

Finally, the fourth main uncertainty is the temporal variability of water fluxes in soil, vegetation and atmosphere. Here, a number of methodologies have been developed to measure components of evapotranspiration such as sap flow measurements and using existing water (hydrological) models. An additional uncertainty is that incident climatic data is often taken a short distance away from the studied stand. In the case of the Belgian NORM site this is from a 24 m-high met tower at SCK-CEN, 15 km away.

Therefore, the study of a forest-type vegetation and of its functioning is opportune and justified to mirror the climate-site-vegetation interactions and to model their impact on radionuclide biogeochemistry.



Figure 3. Heterogeneity in the soil at the Belgian NORM site.

5.1.3 Approach employed to analyse uncertainties and main outcomes

The soil-to-tree transfer factor is a compound parameter comprising in a non-explicitly way a series of processes that are time-dependent, and as such, it is a very crude approximation to understand transfer. The main “hidden variables” that are not considered in transfer factors, and lead to a high degree of uncertainty in them, are (a) effects of vegetation development on the water and element cycling, (b) the underground contribution to the water uptake by vegetation and (c) the vegetation role as a sink and possible vector of long term radionuclide recycling and accumulation (in other words, the transfer factor is likely not constant over long periods of time). These variables are often neglected due to a lack of adequate data. In particular, water fluxes need to be measured in parallel with the biological cycle of radionuclides, and this is not usually done, except where there is a dedicated observatory site, such as the case of the Belgian NORM site used in the TERRITORIES project. In addition, representing transfer by a constant transfer factor assumes that the transfer to plants is a linear process, which is not necessarily in agreement with reality, so the use of transfer factors has an important conceptual uncertainty.

The process adopted in the present study to isolate uncertainties in sampling and monitoring, with particular regards to investigating the soil-to-plant transfer, was:

- (a) To characterise, both in quantitative and dynamic terms, stand eco-physiological compartments which determine water fluxes, i.e. stand leaf area index (LAI), tree dimensions and rooting depth.
- (b) To estimate the main terms of the hydrological cycle in forest stands (soil water content, water table level, stand transpiration and interception) and therefore identify the sources of water used by trees (precipitation vs groundwater).
- (c) To characterise the main soil hydrological variables: porosity, hydraulic conductivity (HC), field capacity (FC), residual water content (θ_R), wilting point (θ_W) and saturation point (θ_S). These variables

are critical as they determine the status of available water (and thus ground-based pollutant elements) to vegetation. Yet, they have important uncertainties because they are difficult to measure (and there are important protocol differences when conducting experiments in the laboratory to measure them), so often they are determined indirectly from semi-empirical equations which are based on certain soil properties such as the fraction of sand, silt and clay, or derived in whole or in part from data acquisition and literature reviews.

(d) To understand the transport of elements in the soil and vegetation by identifying the key parameters, which again carry the most uncertainty: the distribution coefficient (K_d), which determine the retardation of element fluxes in relation to soil water fluxes, and the selectivity coefficients (S_c), which determine the retarding of solutes with respect to water across the plant xylem and phloem vessels.

The K_d uncertainty is a special case here, as the uncertainties are elevated due to high spatial variability (two samples taken close together can have a very different K_d), sampling (extractable soil solution is notoriously difficult to extract from highly compacted soils) and instrumental variability (different techniques e.g. in-situ measurement, batch extraction or column extraction techniques can lead to different K_d as the various operationally-defined extractable and non-extractable fractions can vary for the different methods). Model-derived K_d s come with their own uncertainties. In the simplest case a generic K_d may not be applicable as it may not capture the characteristics of the site (for example the Belgian NORM site soil is in reality a chemical mixture embedded in a CaF_2 matrix), whilst a geochemically modelled K_d (e.g. calculated using PHREEQC code) requires many geochemical parameters and not all of them are available, so again “generic” data may have to be used. An intermediate solution, such as a parametric K_d approach as a function of sand, silt, clay, pH and other basic variables, does not require so many parameters, but still has large uncertainties and potential difficulties if some of the actual soil properties exceed the range of applicability of these empirical functions.

The S_c has also significant uncertainties due to the fact that it may have to be estimated using chemical analogue approaches when direct data for the radioelements necessary is not directly available due to expense of measurements, and it has to be model-derived. However, these uncertainties are more controllable when the object of interest is the soil-to-whole tree transfer factor, because the main driving force is the S_c regulating transfer from soil to roots which can be measured directly.

(e) A forest plot was also instrumented with sap flow sensors, light sensors, soil moisture probes, piezometers, rain gauges, temperature sensors, etc. integrated in an electronic data logger (Figure 4). These sensors detect time-variations in the system critical to forest functioning and radionuclide cycling, such as temperature, precipitation and evapotranspiration cycles across the day/night cycle and the seasonal cycle. Here, the main source of uncertainty is instrumental siting and calibration. For example, to convert sap flow sensor voltages to flow data, an instrumental calibration is necessary and this has its own uncertainties.

(f) To monitor seasonally samples of soil, roots, bark, wood, branches, tree needles and litter fall for radionuclide determination in order to provide points of validation for modelling. Here, an important source of uncertainty is the limited amount of samples that can be collected and analysed for transuranic elements due to the financial cost of these analyses.



Figure 4. Monitoring instrumentation at the forest of Belgian NORM site.

5.1.4 Conclusions

The study performed at the Belgian NORM site shows that the largest uncertainties in sampling and monitoring lie in the experimental determination of H_C , F_C , K_d and S_c , determining as they do the maximum extractable water (EW) for trees in the rooted zone. For the K_d in particular, the main difficulty was that for the highly-impermeable sludge soil present at the Belgian NORM site, pore water is extremely difficult to extract. Therefore, laboratory batch experiments are planned, as well as using geochemical modelling (PHREEQC) to calculate theoretically K_d s based on the already available detail data for the soil mineralogical and chemical composition.

At the time of the present deliverable (D9.60), a significant advance with the approach chosen in the Belgian NORM site has been done, which was to explore the uncertainties of variables affecting radionuclide transfer to vegetation during the process of optimisation of operational integrated vegetation-soil-water models for the Belgian NORM site biosphere, including a detailed representation of the hydrology of the site. The focus is on a key output of the model: a dynamically calculated transfer factor, allowing the opportunity to make a qualitative, science-driven sensitivity analysis of the key variables affecting it, as well as factorising its temporal variability. In the near future the models will be compared and the model and data uncertainties will be qualitatively analysed, drawing conclusions and making recommendations on how to address (and hopefully reduce) these uncertainties.

5.2 A case study: Uncertainty quantification in airborne radiation measurements, in the Fukushima post-accidental monitoring program

5.2.1 Background

In long-distance distribution of radionuclides over the territory, e.g. following nuclear accidents, airborne gamma-ray measurements provide valuable monitoring data for reconnaissance investigation and inventory estimations (Sanderson et al., 2001). Flight-line pattern (direction and spacing), height and speed should be designed according to the background radiation, target radiation, geometrical shape and size (Minty, 1997; Kass, 2013).

In Fukushima post-accidental situation, the Japanese Ministry of Education, Culture, Sports, Science and Technology (MEXT) performed periodical airborne radiation campaigns to monitor temporal evolution of contamination over the Fukushima region (surface soil is targeted), which the data are available to the public through <https://emdb.jaea.go.jp/emdb/en/>.

5.2.2 Main sources of uncertainty

Airborne radiation measurements contain two major uncertainty sources: (i) lack of data between the flight-lines and (ii) volumetric mechanism of geophysical measurements.

Flight-line spacing. Since acquired airborne measurements are denser along the flight-lines, the flight path is usually designed parallel to azimuth of maximum spatial variability (if geometrical anisotropy exists and is known). In addition, flight-line spacing must be as small as not to alias the reality, i.e. providing unrealistic results or missing local hot spots between the lines (Reeves, 2005). The effect of flight-line spacing on the estimation of cesium-137 inventory in the soil was quantified in NW England and SW Scotland (Sanderson et al., 2008): using subsampling technique, i.e. increasing flight-line spacing from 50 m to 500 m (10 km × 10 km area) and from 500 m to 5 km (50 km × 50 km area), the general outlines of depositional area were preserved, and the estimated inventory differed less than 10%. However, local spatial variabilities were not distinctly marked by sparser surveys.

Flight-line spacing is in fact a compromise between the survey costs (financial, time delay, sampling limitations, etc.) and monitoring objectives and targets, e.g. level of contamination, waste volume, desired resolution, etc. (Desnoyers and Dubot, 2014a, 2014b). In regional or national scale inventory investigations, the flight-line spacing is suggested to be one kilometre or greater, while between 50 and 400 m is recommended for detailed mapping projects. In searching radioactive sources or individual field boundaries, flight-line spacing of less than 50 m is proposed (IAEA, 2003).

Volumetric measurements. Gamma radiation detection is a sort of averaging over a volume of investigation. Considering a source of gamma-ray on the ground, emitting gamma-rays with the intensity of $1 \text{ nSv}\cdot\text{s}^{-1}$, the radiations might be detected several meters away of the source. Just above the source, at the height of 1 m, the detector shows just lower than $1 \text{ nSv}\cdot\text{s}^{-1}$, because of stochastic mechanism of gamma-ray detection and the 1 m vertical distance from the source. In addition, emitted gamma-rays from the source could be detected in surrounding areas, where no source is beneath the detector (Figure 5). Therefore, indicating the exact location of the radiating source is uncertain using a gamma-ray detector instrument. In addition, natural randomness of detected value due to stochastic mechanism of detection adds more uncertainty to gamma-ray detection surveys. In sum, volumetric

measurements: (i) underestimate the source radiation at the source location, (ii) detect radiation in locations, other than the source and (iii) are uncertain in identifying source location.

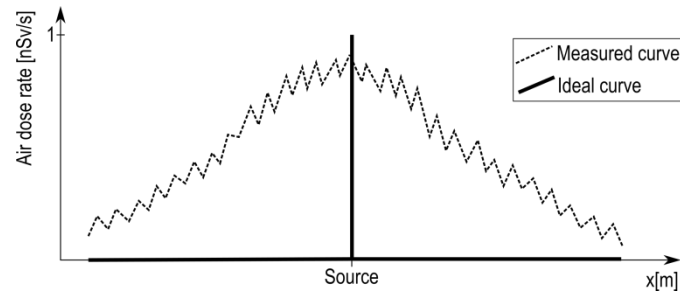


Figure 5. Comparing Dirac-shape ideal curve of radiation source versus measured air dose rate curve, which is a volumetric measurement.

The airborne measurements are sort of volumetric measurements that the measured value is attributed to a circular area on the ground, beneath the detector, called Field Of View (FOV). For the Fukushima surveys, radius of the FOV is suggested to be considered equal to the detector height, i.e. 350 m (Lyons and Colton, 2012). According to Malins et al. (2015), more than 75% of recorded gamma-rays, at the height of 300 m, are originated from the radius of less than 350 m (Figure 6).

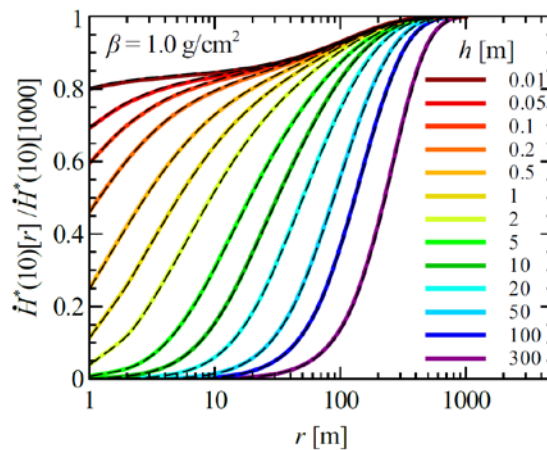


Figure 6. The FOV of soil radiocesium at various heights above the ground (Malins et al., 2015). Vertical axis is the fraction of detected gamma-ray from a specific horizontal distance. It is considered that all the detected gamma-rays are emitted from the distance of less than 1 km.

Volumetric Nyquist frequency was proposed to combine these two uncertainty sources, i.e. sampling rate or flight-line spacing and volumetric nature of measurement, and to provide a quantification value of data for the volumetric discrete samples (Masoudi et al., 2017).

$$f_{Nyq}^{vol} = \frac{1}{SR + D_{FOV}}$$

where volumetric Nyquist frequency (f_{Nyq}^{vol}) is the inverse of sum of sampling rate (SR) and diameter of the FOV (D_{FOV}).

If a target is larger than the summation of sampling rate and diameter of the FOV, it could be characterized by the survey; if not, the probability of its characterization would be in proportional to volumetric Nyquist frequency of the survey.

5.2.3 Approach employed to analyse uncertainties and main outcomes

Fukushima airborne radiation surveys. From June 2011 to December 2013, the MEXT conducted several airborne measurement surveys on a semi-circle with 80 km radius centred at the Fukushima Daiichi Nuclear Power Plant. Japanese Atomic Energy Agency (JAEA, 2014) converted the counting rates at the flying altitude of ~350 m into the deposits estimated at ground surface, in three steps: (i) altitude correction to 1 m above ground level; (ii) conversion of count rates into dose rates; and (iii) conversion of dose rates into ground surface activity of cesium-134 and cesium-137, in [Bq.m⁻²] (Gonze et al., 2014; JAEA, 2014).

Sampling rate along the flight-lines is about 40 m, and perpendicular to the flight-lines, i.e. flight spacing, varies from 600 m to 2,000 m, depending on the area and the airborne survey. Six squares of the length 20 km were chosen as test sites (T1 to T6 on Figure 7), representing a variety of geographical, land-use and radiocesium deposit range.

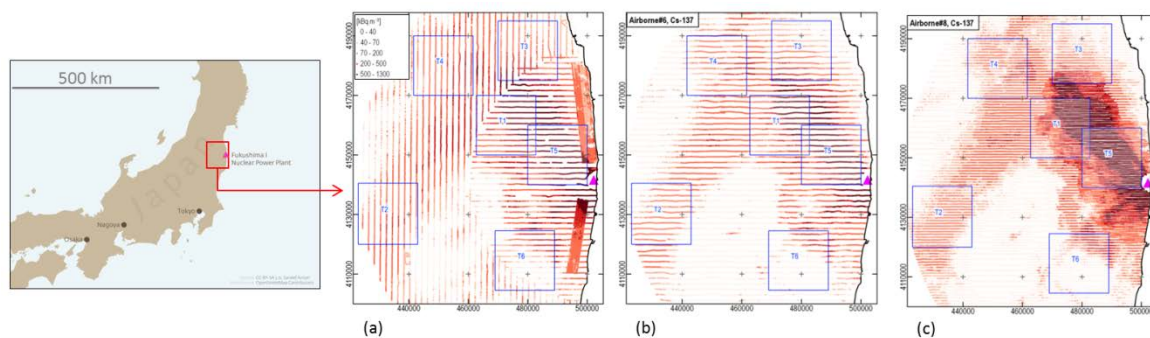


Figure 7. Airborne surveys #4 at November 2011 (a), #6 at November 2012 (b) and #8 at November 2013 (c) over the Fukushima region. The six test sites (T1 to T6) are also illustrated.

Characterization sensitivity to flight-line spacing. In the context of the TERRITORIES project, Masoudi et al. (2018 and submitted) quantified soil contamination characterization error of airborne survey #8, by applying geostatistical estimators to different selections of flight-lines of increasing spacing. At first, point ordinary kriging estimator was applied to the flight-line selections to calculate the punctual error (Table 4). Then, block ordinary kriging estimator was applied to the datasets in order to produce contamination maps (Figure 8), and to classify the area due to contamination threshold. Comparing the classifications with the densest flight-line spacing (Figure 8a), classification error was calculated for sub-sampled datasets (Figure 8b-g, Table 4). Both punctual and classification errors are highly correlated with the flight-line spacing ($r^2 > 0.78$, Figure 9): depending on the test site, increasing flight-line spacing for one kilometre increases the errors from 3% to 9%.

Based on these regression error models, the punctual and classification errors for each test site are calculated for the other airborne campaigns (Table 4).

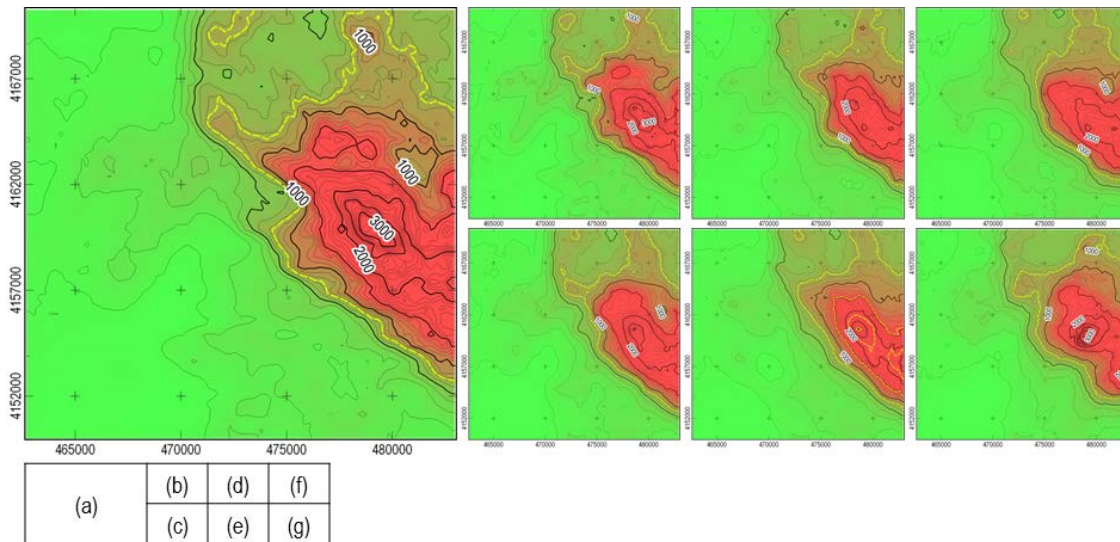


Figure 8. Contamination map of cesium-137, estimated by block kriging at the grids of 250 m × 250 m in test site T1, using all flight-lines with flight-line spacing of 920 m (a), sub-sampled flight-lines with flight-line spacing of 1,270 m (b), 1,840 m (c), 2,530 m (d), 2,890 m (e), 3,380 m (f) and 4,050 m (g). The contour values are in [kBq.m⁻²]. Dashed-line is contamination threshold, i.e. 732 [kBq.m⁻²]. Geostatistical calculations are done by Isatis (Geovariance) commercial software, and the map was realized by QGIS Girona open-source software application.

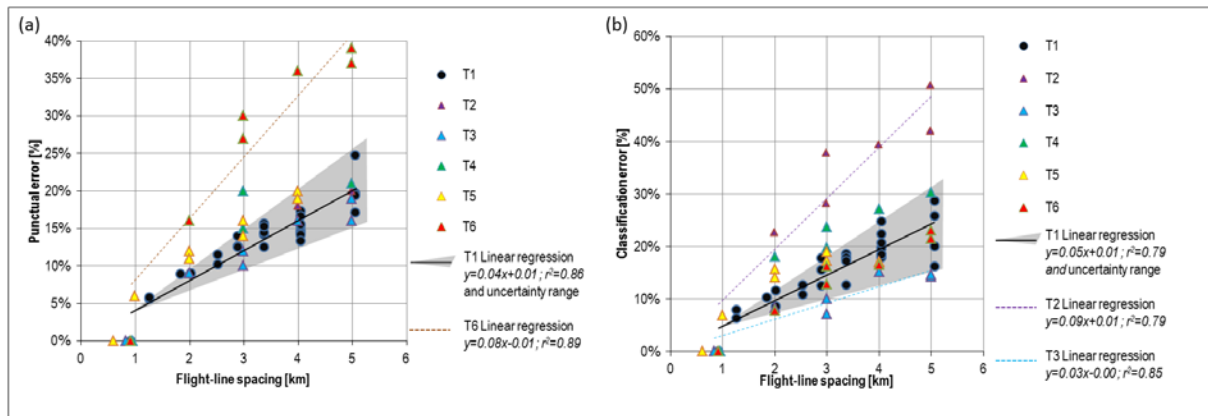


Figure 9. Punctual (a) and classification (b) error, sensitivity to flight-line spacing.

Table 4. Flight-line spacing and related specifications in the test sites. The errors were generalized to the previous airborne surveys (#3 to #7).

Test site	Airborne	Base date	Flight-line spacing [km]	Volumetric Nyquist frequency [km ⁻¹]	Punctual error	Classification error
T1	#3	02.07.2011	1.8	0.40	8%	10%
	#4	05.11.2011	1.8	0.40	8%	10%
	#5	28.06.2012	1.8	0.40	8%	10%
	#6	16.11.2012	1.8	0.40	8%	10%
	#7	28.09.2013	0.6	0.77	3%	4%
	#8	19.11.2013	0.6	0.77	3%	4%
T2	#3	02.07.2011	2	0.37	9%	19%
	#4	05.11.2011	2	0.37	9%	19%
	#5	28.06.2012	1.8	0.40	8%	17%
	#6	16.11.2012	1.8	0.40	8%	17%
	#7	28.09.2013	0.9	0.63	5%	9%
	#8	19.11.2013	0.9	0.63	5%	9%
T3	#3	02.07.2011	2	0.37	9%	6%
	#4	05.11.2011	2	0.37	9%	6%
	#5	28.06.2012	1.8	0.40	8%	5%
	#6	16.11.2012	1.8	0.40	8%	5%
	#7	28.09.2013	0.9	0.63	5%	3%
	#8	19.11.2013	0.9	0.63	5%	3%
T4	#3	02.07.2011	2	0.37	9%	11%
	#4	05.11.2011	2	0.37	9%	11%
	#5	28.06.2012	1.8	0.40	8%	10%
	#6	16.11.2012	1.8	0.40	8%	10%
	#7	28.09.2013	0.9	0.63	5%	6%
	#8	19.11.2013	0.9	0.63	5%	6%
T5	#3	02.07.2011	1.8	0.40	8%	10%
	#4	05.11.2011	1.8	0.40	8%	10%
	#5	28.06.2012	1.8	0.40	8%	10%
	#6	16.11.2012	1.8	0.40	8%	10%
	#7	28.09.2013	0.6	0.77	3%	4%
	#8	19.11.2013	0.6	0.77	3%	4%
T6	#3	02.07.2011	1.8	0.40	13%	10%
	#4	05.11.2011	1.8	0.40	13%	10%
	#5	28.06.2012	1.8	0.40	13%	10%
	#6	16.11.2012	1.8	0.40	13%	10%
	#7	28.09.2013	0.9	0.63	6%	6%
	#8	19.11.2013	0.9	0.63	6%	6%

Airborne dataset vs. soil data dataset. Following Fukushima nuclear disaster, several data gathering campaigns surveyed the radionuclide propagation in the environment. However, the acquired datasets do not have the same sampling dimension. For example, the airborne measurements are some sort of averaging over a circular field of view, beneath the sensor; while the soil analyses are much more punctual. Masoudi et al. (2019) tried to find out to what extent (and in which aspects) soil and airborne datasets carry the common information. Addressing this problem conducts to better understanding of the datasets, in order to achieve a more accurate contamination characterization.

Each dataset has its own negative and positive points, and a wise solution to reduce the drawbacks while benefiting of the advantages worth being checked (e.g., Wainwright et al., 2017). Since the residuals are correlated, co-kriging algorithms could be used in order to integrate these datasets (Masoudi et al., 2019). Being direct and punctual makes the soil data as a reference of estimation, i.e. the primary variable in the co-kriging algorithm. The airborne data could be used as secondary variable, which helps in improving the co-variogram (because of good coverage and high spatial correlation).

5.2.4 Conclusions

Airborne measurements and soil analyses are complementary data for soil contamination characterization in a post-accidental situation. Soil analyses provide precious information whereas the chain of sampling design, terrain missions for taking and bringing samples to the laboratory and analysing them, demands time. On the other hand, airborne data are very quickly available in a large scale, which is suitable for rapid decision-making over vast contaminated areas.

Here, the focus is on the error of characterization by airborne measurement as a function of flight-line spacing, which should be designed according to the characterization importance. Denser the flight-lines, lower the characterization error. In general, the characterization error increases 5% by adding one kilometre to the flight-line spacing, whereas depending on spatial continuity of the measurements, this percentage could be changed from 3% to 9%.

The next step could be evaluating socio-environmental consequences of the characterization error. It helps drawing a clearer vision of the importance of flight-line density, hence decision-making based on more tangible facts.

5.3 A case study: Uncertainty in the conceptual scheme of processes occurring in a Polish lake displaying NORM

5.3.1 Background

Extraction operations are known to increase the transfer of pollutants to the surrounding environment and the present challenge is to develop green mining strategies. It concerns not only the production stage but also remediation operations and associated waste management. Many questions arise from past activities, especially regarding the effectiveness and the long-term sustainability of the remediation works. Among these questions is the fate of diffusive and advective transport of pollutants at redox interfaces (Poinssot and Geckeis, 2012).

Radium is one of the specific contaminant of NORM industries, including uranium mining, coal activity and oil and gas production. Co-precipitation of radium (Ra) with barium (Ba) as radiobarite is a usual approach to remove Ra from industrial wastewaters. Barite is considered as one of the main Ra-bearing phases in natural and anthropogenic systems. Although barite is frequently cited as a highly insoluble phase under oxidizing conditions, it is also suspected to be dissolved under sulphate-reducing (Baldi et al., 1996; Landa et al., 1986; Bolze et al., 1974).

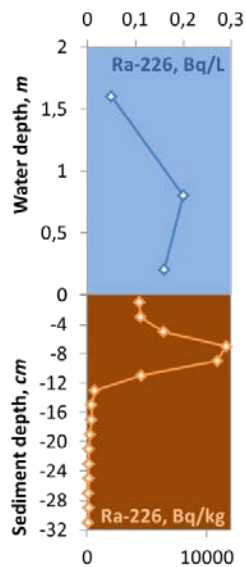
The Upper Silesia Coal Basin is situated in the southern part of Poland and is known as one of the four European radioecological observatories (<https://radioecology-exchange.org/content/upper-silesian-coal-basin>).

Deriving from underground coal mining activities, brines containing high concentrations of Ra (up to 300 Bq L⁻¹) and Ba are mixed with underground waters containing sulphate ions SO₄²⁻ allowing the removal of Ra and Ba from waters by co-precipitation (Chalupnik et al., 2001). As a result, much of the Ra isotopes are trapped into radiobarite (Ba,Ra)SO₄ in the bottom deposits of settling ponds (Bzowski et al., 2015); among them the Rontok Lake was used by the local coal mining industry from 1977 to 2002. The Rontok Lake was since unused and the renewal of surface waters allowed its return into a “wild ecosystem” functioning. The lake and its surrounding environment are presently characterized by abundant aquatic and terrestrial fauna and flora. As such, the Rontok Lake can be considered as an analogue of long-term situation of rich radium waste storage.

5.3.2 Main source of uncertainties

Since 2013 several sampling campaigns were performed by IRSN and GIG (Central Mining Institute, Poland) to study Ra behaviour in the vicinity of former and ongoing coal mines of the Rontok Lake area (Courbet et al., 2016). A specific attention was paid to the hyporheic zone of border streams where groundwater tends to mix with surface waters within the river bed, and in a lesser extent, to the water-sediment interface of the Rontok Lake. Both represent redox interfaces where solid bearing phases of radium (barite, metal oxyhydroxides) undergo various geochemical processes which have not been quantified yet.

In the Rontok Lake, the potential release of Ra through the water-sediment interface was further investigated (Zebracki et al., 2019), as:



- the radium activity concentration was shown to increase from the water surface to the bottom (Figure 10),
- the bottom sediment displayed reducing conditions,
- and the radium activity concentration was significant and higher than expected regarding the local hydrochemical background.

Figure 10: Depth profile of Ra-226 activity concentration in water (Bq.L⁻¹ versus m) and sediment (Bq.kg⁻¹ of dry sediment versus cm).

In the framework of the TERRITORIES Project, further investigations were performed on the Rontok Lake which provides a rare opportunity to investigate the long-term sustainability of Ra-rich sludge storage. Indeed, the development of wildlife constitutes a good analogue to the long term-evolution of storage. Focusing on the identification of unexpected but relevant processes at the water-sediment interface, the present study also aimed at improving the modelling of the pathways of the radionuclides transfer occurring in a freshwater ecosystem.

5.3.3 Approach employed to analyse uncertainties and main outcomes

The study strategy aimed at (1) characterizing the potential of Ra remobilisation within the sediment and its release throughout the water-sediment interface and at (2) investigating the speciation of Ra in the sediment.

A single campaign was performed in April 2018, allowing the sampling of water, sediment and pore water of sediment (Figure 11). Several analyses were conducted in order to determine the vertical profiles of the radionuclides activity concentration, the grain-size distribution, the major and trace elements concentrations. Selective chemical extractions were conducted at IRSN laboratory using the collected sediment samples, and throughout the addition of selective reactants for iron-bearing phases, such as the reagents TAMM and CDB (Othmane et al., 2013).

At the time of the present deliverable, only gamma results were added to the TERRITORIES Library Database (CONCERT-TERRITORIES D9.59, 2019).

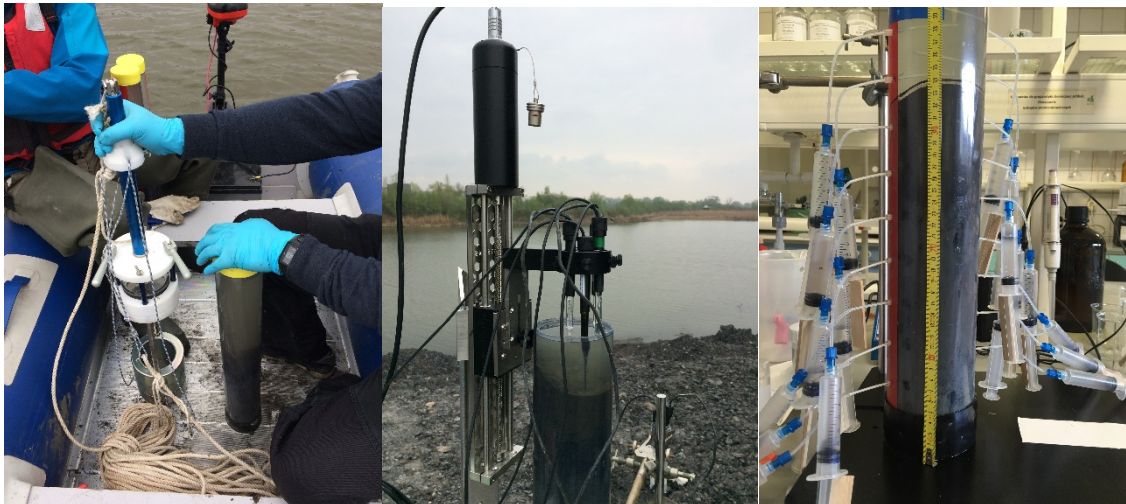


Figure 11: Pictures of the sediment corer (left), the micro-electrodes measurement device (middle) and the pore water of sediment collection device (right).

In the Rontok Lake, the rapid consumption of the oxygen within the top millimetres below the water-sediment interface was evidenced (Figure 12), implying the establishment of reducing conditions due to an active bacterial degradation of the freshly deposited organic matter (early diagenesis process). The sulphide production observed in the deepest sediment evidenced sulphate-reducing conditions (Figure 12).

The analysis of Ra concentration in the pore water of sediment was performed by ICP-MS but the very low level of Ra coupled to the small volume samples (< 6 mL) didn't allow its quantification (i.e., Ra-226 below 0.2 Bq.L^{-1}). In order to achieve low level Ra analysis in small volume samples (0.2 mL), new analytical developments are currently carried out in our laboratory (Verlinde et al., 2019).

Contrary to Ra, Ba is an element which is much easier to measure and serves as Ra analogue in environmental studies (e.g. Landa, 2007). Based on previous data obtained in the Rontok Lake, the high correlation observed between Ra and Ba in the particulate fraction of bottom sediment confirmed the potential of Ra trapping by Ba phases. In the pore water of sediment, the gradient of Ba concentration observed just below the water-sediment interface evidenced the remobilization of Ba (Figure 12). Once Ba is remobilized it might be released by diffusion throughout the water-sediment interface. Regarding the surface water, then the bottom sediment becomes a source of Ba throughout diffusive processes, although being a trap in the particle fraction at the same time.

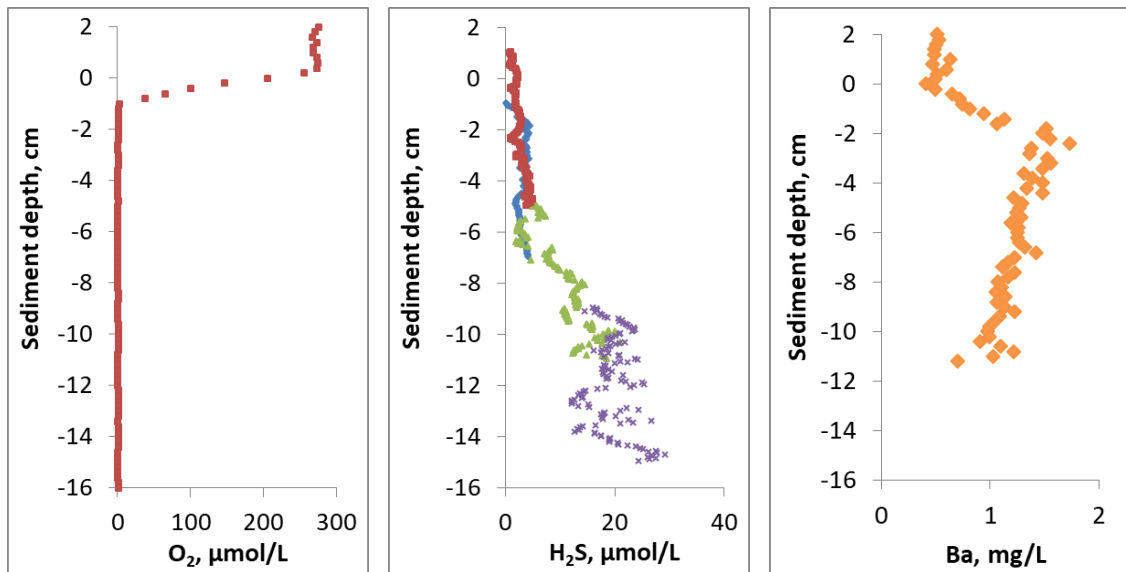


Figure 12. Depth profiles in the pore water of sediment of the concentration of dissolved oxygen (left), sulphide (middle) and barium (right). The water-sediment interface corresponds to the depth 0.

The speciation study evidenced that the fraction of Ra associated to carbonate minerals - and to a lesser extent to iron/manganese oxides - varied along the sediment depth (i.e., 27, 4 and 8 % in the deep, middle and surface sediment deposits, respectively). Although the fraction of Ra associated to sulphate minerals is not known at the present time, barite should be the main Ra-bearing phase in surface and middle Rontok sediment deposits. However, the obtained results showed that Ra is not only trapped within the bottom sediment in the form of radiobarite but also in the form of carbonates minerals and less crystallized phases such as iron and manganese oxyhydroxides, which are considered more reactive regarding the environmental processes.

5.3.4 Conclusions

Future radioecological issues emerge from the functioning of the aquatic systems storing NORM bottom deposits once they return back to a wild state. In the Rontok Lake, the occurrence of early diagenesis process was discussed regarding the potential of bottom sediment as being a delayed source of contamination for the surface water. The primary objective of our study was to reduce the uncertainty related to the modelling of the radionuclides transfer by improving the knowledge on the transfer pathways.

In the near future, a fit-for-purpose model will be designed in order to determine whether the diffusive transfer of Ra throughout the water-sediment interface has consequences regarding the water quality, and if such process must be integrated in the long-term evaluation of sustainability of Ra-rich sludge storage under water cover.

6 CONCLUSIONS AND GUIDELINES

Levels of radioactivity concentration in the environment compartments (for instance in soils and water) are not only variable with time, due to several processes, including radioactive disintegration, migration and dilution of radionuclides, but also in space, mainly because the initial radioactivity is not homogeneous in the contaminated areas, but also due to other aspects as the characteristics of the area (e.g. soil density, porosity or chemical composition of the soils), which will affect the movement of the radionuclides and can be very variable with space.

To make decisions, non-measurable quantities such as effective dose for humans are used. Those quantities are calculated by using real measured values of physical quantities, which should be properly quantified. Activity concentrations in soils or vegetables, or ambient dose equivalent are examples of those measurable quantities. How to properly determine correct values to those measurable quantities is one of the main concerns for assessors and for decision makers. Moreover, for many applications the values are not measured, but estimated using available models. Obviously this determination possesses a degree of uncertainty, and the possible uncertainties and variabilities which can affect the final result should be also properly determined. Many of the possible sources of uncertainties and variabilities have been extensively addressed either in this project (CONCERT-TERRITORIES D9.62, 2019) or elsewhere (e.g. GUM, 2008). Definitions on some of the terms used in this guidance (e.g. uncertainty and variability) are given below. One of the sources of uncertainty, well-known and treated in other fields, as in chemical laboratories (cf. Eurachem, 2007), and also in radioactivity measurements (cf. ICRU, 2006), is often not quantified as a part of the measurement process in radioecology. This is the so called sampling uncertainty.

To calculate sampling uncertainty many aspects should be considered: the temporal and spatial variations, but also the size of the sample, either in terms of mass or volume when dealing with sampling of soils or waters, either in terms of collection time when dealing with direct ambient dose equivalent, counts per second (cps), or any other quantity affected by size. The intention of this document was not to create new methods to address this specific problem, but to compile all available information and include the own experience in the TERRITORIES project to provide guidance to other scientists.

One of the problems included in this deliverable D9.60, under different points of view, is how to define optimum locations for monitoring and sampling. Also the optimum number of samples necessary to adequately characterize the variability of the contaminations in a given time and area has been addressed. Some of the methods are based on the judgement of experts, who define a-priori what should be measured, where should the samples be taken and what the size of the samples should be. Some other methods are focused on how to statistically define the optimum locations and the number of samples to be taken based, among other parameters, on the level of contamination in a given location. None of those methods are perfect for every situation, and many times combinations of the methods are necessary. For instance, a purely random sampling can detect by chance every hot spot in a contaminated place, but it can also skip them. A systematic stratified method would provide a general idea of where the locations of the more contaminated zones are. But still there are specific problems, as for example those related only with hot-particles that require a further refinement.

Usually the specific problem of defining where samples should be taken, in view of the existence of hot particles or hot spots, is approached in a multi-stage process:

1. The first stage broadly identifies where the contamination is expected by using expert judgement. This is often based on previous experience or information, or on the outcome of dispersion models. This first stage can clarify whether hot particles are expected to be correlated with zones of high activity levels or not. For instance, the release of hot particles in Sellafield cannot be completely related with the release of other contaminants dissolved in the water. On the contrary, the location of the initial contamination at the Belgium NORM site is well defined and hot particles are not expected. This first stage can cause problems if the initial information is not sufficient.
2. The second stage will use any of the methods mentioned in this guidance to define the location of all the samples required, within the area previously defined. Also the number of samples and their size (e.g. in terms of mass, volume or time) should be specified in this stage. Random, stratified or adaptive cluster sampling are examples of these methods.
3. The third stage will refine the initial characterization of the affected zone by using the quantities previously monitored. Usually different methods are used in this stage, for example making quick ambient dose equivalent measurements (by foot, car or plane) to provide a first general view, or performing continuous measurements using alpha detectors very close to the soil, as done in the case of the beach close to Sellafield. Also the collection of samples (e.g. of sand, sediments or water) in defined locations is part of this stage. In this stage as many refinements as necessary should be performed. For example, if hot particles can be related with the appearance of zones with higher contamination, more exhaustive monitoring should be performed in those more contaminated zones, while less effort is required in the less contaminated zones.

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4. A fourth stage interpolates all the measurements performed in the previous stages and in laboratories, to provide quantitative results in the entire affected zone. Kriging methods, for instance, are used for a multi-dimension interpolation, as was shown in the case of Fukushima.

This process will provide an acceptable initial characterization of the contamination in the zone, although, as it has been previously pointed out, several aspects should be at least considered and discussed.

A first aspect to consider is related to the **repeatability of monitoring process**. A sample or in-situ measurement cannot be repeated under exactly the same conditions. For instance, if a sample of soil is taken to make laboratory measurements, a second sample of soil at the same location will be only approximately the same, but it cannot be exactly identical. The same will happen with airborne measurements, especially when the half-lives of the radionuclides of interest are very short. In this case, a second survey after some months will be very difficult to be done at exactly the same points

and the measurements will be different because of several processes (e.g. radioactive decay or migration due to rainfall). Some effects can be taken into account by mathematical corrections, as for example corrections for radioactive decay.

A second aspect to consider is the **fractal character of the contamination** (see Figure 13). When using higher and higher resolutions to characterize the radioactivity in a zone, a similar contamination pattern can be observed due to unavoidable variability. The contamination pattern is in principle smaller when the resolution is higher, but with a very similar shape of the contamination pattern. This effect can be visualized in Figure 14, which shows the effective dose due to natural sources in different size populations.

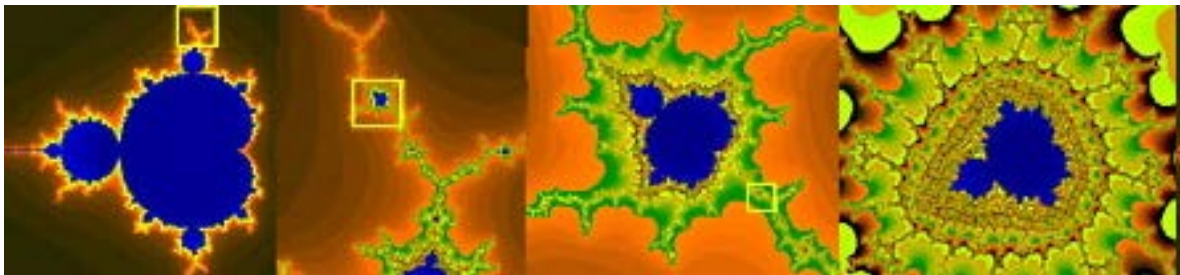


Figure 13. Mandelbrot fractal. This often used fractal can show how a higher resolution in the measurements will provide the same degree of complexity in the characterization than a lower resolution.

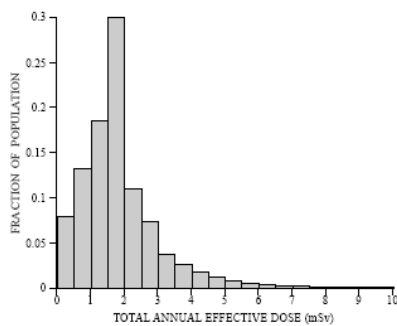


Figure XV. Distribution of population of fifteen countries with respect to total annual effective dose.

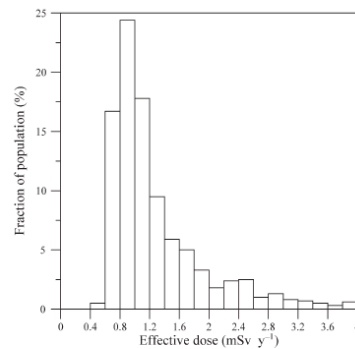


Figure 4. Distribution of population with respect to effective dose from natural radiation sources in Spain.

Figure 14. In the left, total annual effective dose of a population of 15 countries (UNSCEAR, 2000). In the right, the same quantity characterized for 1 country (Spain) (García-Talavera et al., 2007).

This problem continues even in the scale of a laboratory analysing samples. For example, the measurement of several 1 kg soil samples, taken at the same location, will provide a distribution of the measured quantity, which will be characterized by the expected values of that quantity (usually average and variance). However, if one of those samples is used to take for instance 10 aliquots of 1 mg to perform alpha spectrometry, the same pattern will be expected. Moreover, the possibility of finding hot particles increases in those small samples.

And this problem continues if additional dimensions are included. For instance, creating 2D maps of contamination in a given space doesn't imply that the contamination in depth (3D maps) will be known.

A third point to solve is the **movement of the radionuclides in the environment during a period** (usually months or years). A survey campaign by means of in-situ measurements (in-situ gamma spectrometry, or ambient dose equivalent), or by sampling material from the contaminated area, and interpolating the measurement results, will provide a view of the observed quantity at that given point in time. However, many processes will affect the concentrations of environmental media with time. Time dependence is usually modelled and sometimes the validity of the models is checked by repeating the measurement campaign at different points in time. Some of the time-dependent effects are well known and can be easily corrected, as the radioactive decay. The uncertainties arising from sampling and monitoring should be included in the uncertainties propagation of the models. Suitable methods for simple cases were treated in another TERRITORIES deliverable (D.9.62). In more complex cases, e.g. when applying geostatistical methods for interpolation, the process for including those uncertainties can be more time consuming, but the scientific basis is well established.

In summary, this deliverable describes and explains some of the methods proposed in the bibliography to address and discuss sampling uncertainty. Many authors state that sampling uncertainty is often the most important contributor to the total uncertainty of the measurement process, especially in environmental sciences. Nevertheless, little attention has been paid to properly characterize it, neither when designing in-situ measurement campaigns nor when defining sampling criteria to reduce the associated uncertainty as much as possible, i.e. to properly characterize variability at different spatial scales. There are methods to quantify this uncertainty, like the empirical or top-down approach, repeating the sampling as often as needed, using different sampling instruments, protocols; or the modelling or bottom-up approach, using a predefined model from sampling theory.

Effort should be taken to explain and train laboratory staff in charge of performing sampling and monitoring campaigns in order to implement a method to quantify the sampling uncertainty.

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