Geostatistical characterisation of contaminated metals: methodology and illustrations

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Abstract

Radiological characterisation plays an important role in the process to recycle potentially contaminated metals. It is a platform for planning, identification of the extent and nature of contamination, assessment of potential risk impacts, cost estimation, radiation protection, and management of material (decommissioning, release, and disposal of generated secondary waste).

Key issues in radiological characterisation are identification of objectives, development of a measurement and sampling strategy (probabilistic, judgmental or a combination thereof), knowledge management, traceability, recording and processing of obtained information.

The active culvert at Studsvik has been in use since 1962, and is currently subject for renovation, but some sections will also be decommissioned. For the sections to be decommissioned, a radiological survey has been performed, with focus on the system surfaces.

Combining statistical and geostatistical tools in the toolbox concept allows improving project performance while reducing costs. The tools are demonstrated using the decommissioning project at Studsvik for the active culvert, with focus on the non-viable sections of the culvert where the conventional survey techniques cannot be used.

This paper describes the geostatistical methodology and its benefits at the different characterisation stages aiming for reliable results in line with the data quality objectives.

Introduction

Dismantling and decommissioning of nuclear facilities or remediation of contaminated sites are industrial projects with huge challenges. The most precise knowledge of the contamination state is required. Multiple objectives have to be considered for radiological evaluation:

- average activity levels should be quantified to allow the categorization of surfaces or volumes (sorted into different radioactive waste categories),
- hot spots (areas of small dimension with significant activity levels) should be located,
- source term (total activity) contained in soils or building structures should be estimated,
- radiation protection and other logistic considerations have to be taken into account.

Specifically applied on metal components and structures (reactor vessels, heat exchangers, coolant loops, pipes, storage tanks…), the characterisation step has strong impact on possible reuse and recycling of materials.

Many estimates are essential for the proper management of these projects. Currently, characterization remains relatively empirical. Accumulated approximations often lead to serious consequences that threaten the project's successful completion, for example through over-categorisation or unexpected contamination, leading to unexpected costs [IAEA, 2001].

Radioactive contamination is generally complex and involves numerous parameters: radiological fingerprint, type of contaminated or activated materials, oxidation surface state, and so on. Numerical modelling often turns out to be very difficult. Consequently, the characterization phase should be efficient and the sampling strategy has to be rational. However, investigations also represent significant costs due to data collection with radiation protection constraints in nuclear facilities and analytical costs ranging up to thousands of Euros depending on the radionuclide. Therefore the entire sampling strategy should be optimized to reduce useless samples and unnecessary measures [OECD/NEA, 2013].

The geostatistical approach, which provides consistent estimates and reliable maps, is an appropriate solution for data analysis. In this paper, the methodology is first presented on a real dataset for the characterisation of the active culvert at Studsvik site. Then a synthetic illustration is used for the characterisation of linear structure (1D-grid) that can represent the pipe network in the active culvert.

Active culvert decommissioning

The active culvert at Studsvik was built 1957 – 62, and most of the system has been in continues use to support the nuclear facilities at Studsvik ever since. The current project for the active culvert includes both sections that will be renovated, and sections that will be decommissioned. A radiological survey of the sections of the culvert that shall be decommissioned have been described [Lidar and Strid, 2013]. Survey models in five levels are applied for sections of the active culvert where samples and measurements changes depending on the radiological classification of the section. The models should ensure adequate scope and quality of the survey. The radiological survey contains loose contamination, scintillation measurements, dose rate measurements as well as nuclide specific analysis of the activity content in material samples, and water samples.

The sections of the piping network and active culvert included in the decommissioning plan have been in operation supporting systems for handling of liquid radioactive waste classified as category 3, 4 or 5 (eight categories once used at Studsvik). The sections of the active culvert that shall be decommissioned are of three types:

- Viable whole culvert of concrete pipe sections
- Non-viable half culvert of concrete pipe sections
- Pipes dye-casted in the ground.

Piping in viable and non-viable sections of the active culvert to be decommissioned are:

- Approx. 1030 m category 3 piping
- Approx. 350 m category 4 piping
- Approx. 590 m category 5 piping.

This also includes 170 m pipe dye-casted in the ground of categories 4 and 5. The decommissioning planning includes approx. 64 m viable culvert and approx. 427 m non-viable culvert. For geographic location of the sections of the active culvert to be decommissioned, see Figure 1.

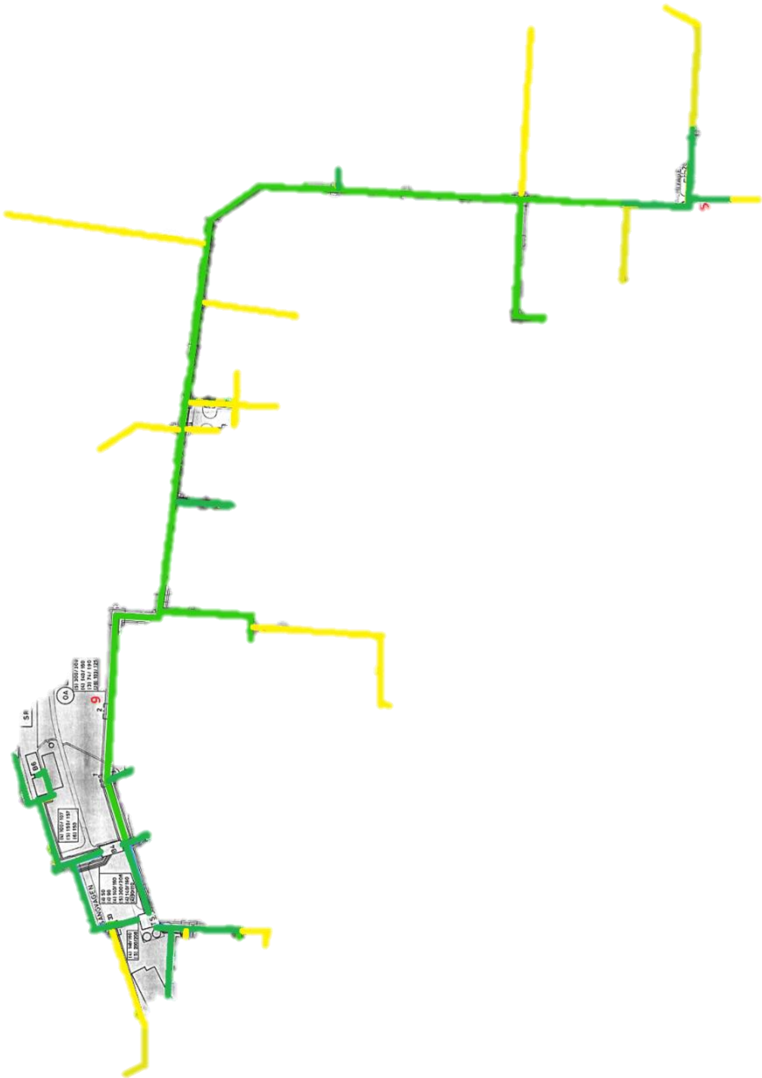

Figure 1. Principal layout of active culvert at Studsvik – remaining sections in green and sections to be decommissioned in yellow (by the time of reporting).

In parallel with the decommissioning project, category 4 piping will be dismantled and replaced. In total 1580 m category 4 piping will be dismantled followed by waste management. Cross-section of a typical non-viable half culvert is shown in Figure 2.

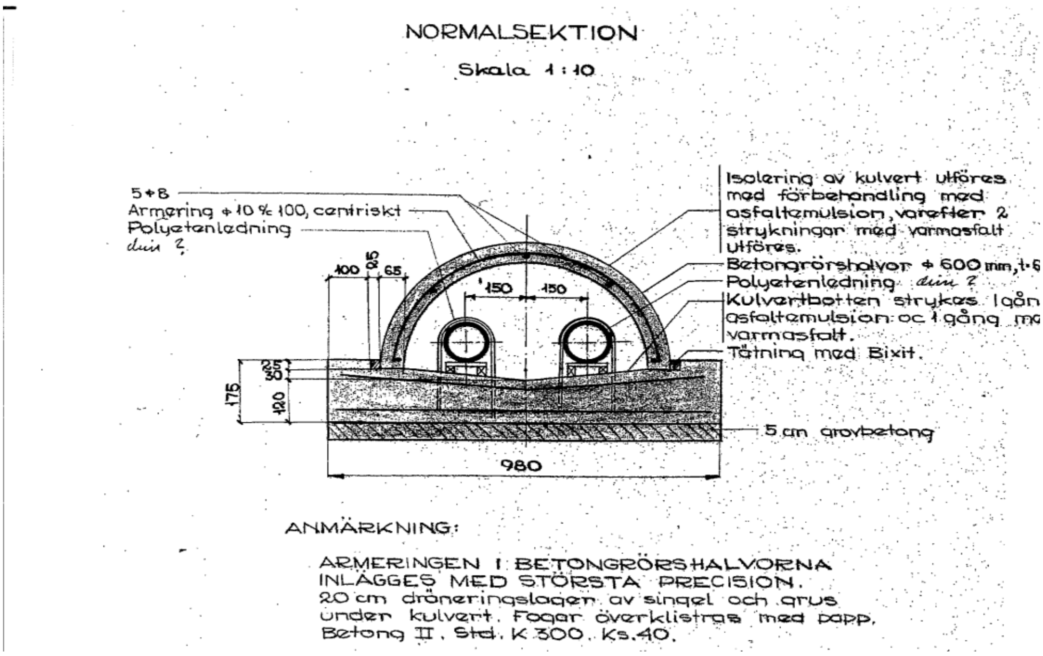

Figure 2. Cross-section of a typical non-viable half culvert.

Along the whole active culvert inspection wells are located, and six wells are included in the decommissioning project. Examples of wells are seen in Figures 3 and 4.

Figure 3. Inspection well with manhole cover and ladder.

Figure 4. Well shaft in two manholes in non-viable part of the active pipes (with and without ladder).

The extent of sections for categories 3, 4, and, 5 planned for decommissioning is seen in Figure 5.

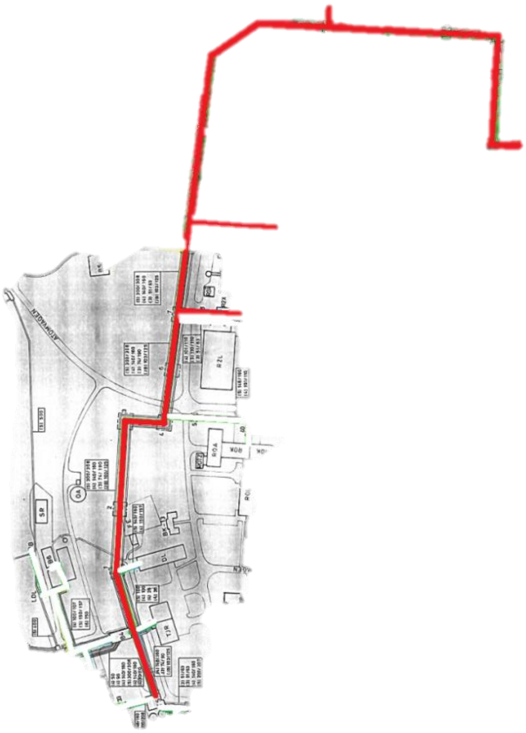

Figure 5. Piping for category 3, 4, and, 5 planned for decommissioning (by the time of report writing).

Controlled area of the active culvert are areas and sections where radioactivity or dose rate over 2.5 µSv/h can exist. The limit values for controlled area is seen in Table 1.

Table 1. Limit values for controlled areas.

Limit values for controlled areas						
Zon			**Non-controlled area**	**Blue**	**Yellow**	**Red**
Radiation zone	µSv/h			< 25	25–1 000	> 1000
Surface contamination	kBq/m^2	α	< 0,4	< 4	4–100	> 100
		β+γ	< 4	<40	40–1 000	> 1000
Air-born contamination	DAC			< 1	1–10	> 10

In the active culvert, the surfaces (on equipment to be replaced or decommissioned) included in the radiological survey (i.e., inner surfaces of the culvert excluding the piping) can be seen in Table 2.

Analysis of results

When all results for a section is available, the analysis of the results are performed. The measured activity is during the project compared with the clearance levels in SSMFS 2011:2 for liquids, material, and buildings for re-use resp. buildings for demolition.

Objective

The objective with the radiological survey of the active culvert is to obtain information about the radiological status in the active culvert. The information is then used for planning for the continued work, radiation protection effort, development of preliminary dose budget, waste characterization and as input to the cost estimation.

Tabell 2. Length in the controlled zone (meter piping of various dimensions) and surfaces (m^2) in active culvert.

Zone	No	NCA+ (m^2)	Blue (m)	Blue (m^2)	Yellow (m)	Yellow (m^2)	Red (m)	Red (m^2)
Culvert wall/roof (viable)			64**	200				
Culvert floor (viable)					64**	86		
Culvert roof (non-viable)			427**	402				
Culvert floor (non-viable)					427**	418		
Piping cat. 3 (top surface)*					1033**	98		
Piping cat. 3 (bottom surface)*					1033**	98		
Piping cat. 4 (top surface)*					355**	305		
Piping cat. 4 (bottom surface)*					355**	305		
Piping cat. 5 (top surface)*			595**	93				
Piping cat. 5 (bottom surface)*					595**	93		
Well shaft	6	75						
Piping support structure*	600					17		
Ladders*	6			19				

+) Non-controlled area
*) Counted as wall or roof surface behind the object.
**) Meter culvert or meter piping of various dimensions.

Experimental

In situ measurements as well as sampling/sampling evaluation has been performed in the culvert sections to be decommissioned. The samplings as well as the in situ measurements have been performed at certain predefined locations.

Sampling

The sampling program covered:

- Water samples

- Material samples (using drilling)
- Smear tests

The water samples were taken as grab samples from different locations in the active culvert. Each water sample was divided into two parts, one part was sent to the radiometric laboratory for analysis, and the other part was kept as reference for potential further investigations.

Material samples were taken using hand held drilling machine and internal working instruction, at different and documented locations based on the survey model. Each material sample (with the highest dose rate) was divided into two parts, one part was sent to the radiometric laboratory for analysis, and the other part was kept as reference for potential further investigations.

Smear tests were taken at each square used for scintillation measurement (see below), by making a cross movement (i.e., like the letter X) between the four corners of the square. The smear tests were analysed in an instrument with ten positions (simultaneous analysis).

Measurements

In situ measurement program covered:

- Dose rate measurements
- Scintillation measurements

The position of the measurements were selected according to the survey model, and documented. The dose rate measurements was performed using a Canberra Colibri instrument and have been electronically transferred to a PC. The frequency of dose rate measurements varies with the radiological survey model, and the results in this report are concentrated to areas with “yellow” surface classification according to [Lidar and Strid, 2013]. Dose rate measurements in this report are taken in the air approx. 1 m above the square where the scintillation measurements were taken, and at the very same time. The dose rate measurement is recorded every time a scintillation result is recorded, and the results are stored in the same data file.

The scintillation measurements have been performed using a Canberra Colibri instrument with a SABG-100 probe. The measurements have been made in squares of 0.5 x 0.5 m marked on the surface to be measured. The whole area of the square has been measured by moving the probe for eight measurements in four rows (in total 32 measurements per square). Each measurement collected an alpha and a beta/gamma value using an integration time of 20 s. The measurements were started with a measurement with the probe in the air approx. one meter over the square to obtain a background value. The results were stored in the Canberra Colibri instrument and transferred to a PC. In total, approx. 1500 alpha resp. beta/gamma measurements were collected in the survey from different squares.

The smear tests were taken in the same squares as above by making an X movement covering the four corners. The smear test measurements were performed in Berthold ten position equipment according to the internal instruction.

Geostatistics

Geostatistics aims to describe structured phenomena in geographic space, possibly in time, and to quantify the estimation uncertainties [Chiles and Delfiner, 1999]. Estimates are calculated from a partial sampling and result in different representations of the contamination, including interpolation mapping (by an algorithm called 'kriging'). But the added value of geostatistics goes much beyond this. Its key feature lies in its ability to quantify estimation uncertainty and provide risk analysis for decision making.

Applied to radioactive contamination, this data analysis and processing framework is recent. However, it has been advantageously employed for more than 50 years by the mining industry for resource assessment [Isaaks and Srivastava, 1989], the oil and gas sector for reservoir characterization [Dubrule, 1998] and extended in recent decades to environmental issues such as hydrogeology, air quality monitoring, conventional pollutants (heavy metals, hydrocarbons), soil science, and so on [Goovaerts, 1997]. It is now increasingly implemented for the characterization of radioactive contaminations in nuclear facilities as well as for contaminated sites and soils [Desnoyers and Dubot, 2011].

Spatial structure and variography

At Studsvik site, datasets have been collected on 0.5 x 0.5 m areas with a 100% surface coverage. 42 areas have been investigated inside the active culvert. Alpha and beta gross rates have been recorded in cps (count per second). For instance, base map of beta values, statistical distribution and correlation with alpha values are presented on Figure 6 for location point #024.

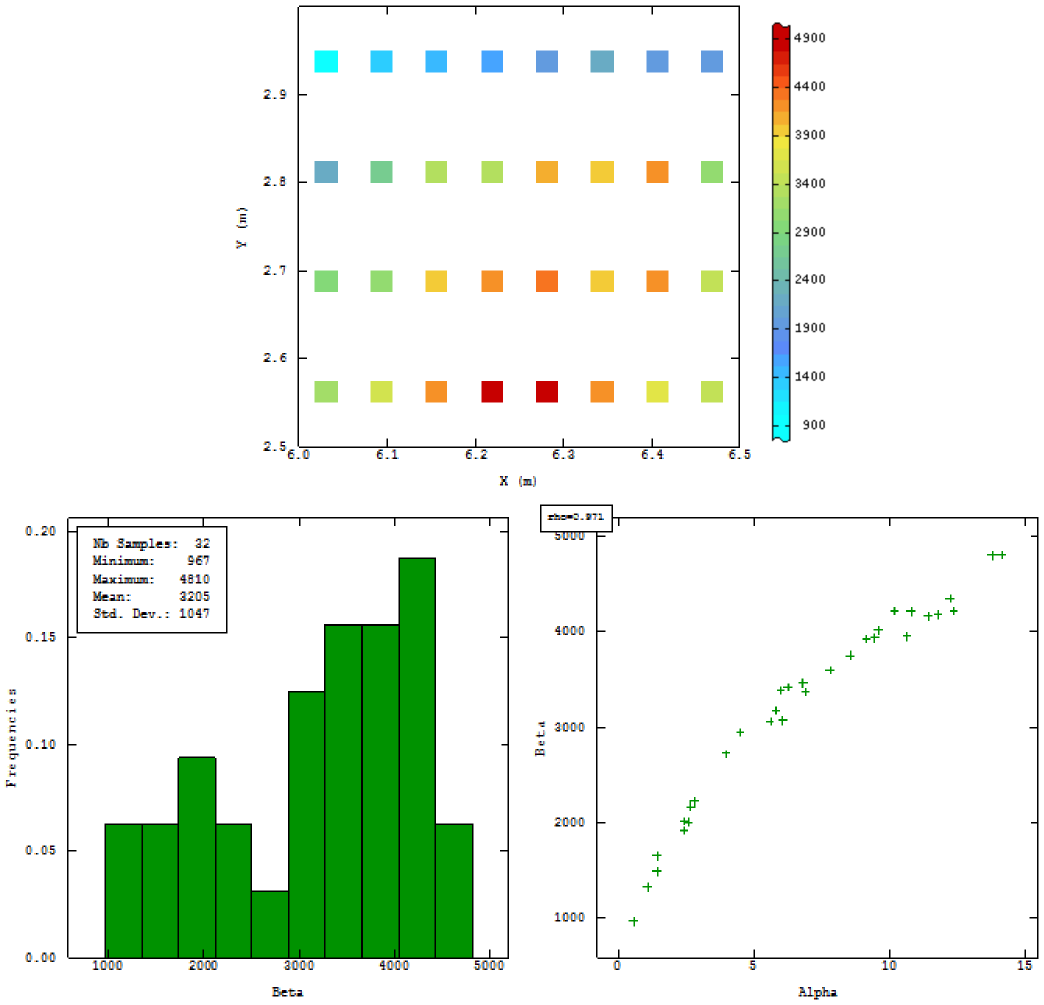

Figure 6: Basemap of beta counts (on the top), histogram and correlation cloud with alpha counts (on the bottom).

This first exploratory analysis indicates a bimodal distribution of beta values (less contaminated versus more contaminated) with possibly two linear regressions with alpha values (regression slope changes around 5 cps). This may be linked with two different contamination events or more reasonably two different deposition behaviours.

Geostatistics assumes the presence of spatial continuity for radioactive contamination. Variability behaviour over distance between data points is the spatial signature of the studied phenomenon. This spatial structure is analysed and interpreted through the variogram which plots the variability between pairs of points (firstly analysed through the variographic cloud). Typically for a structured phenomenon, this variability increases gradually and stabilizes at a certain sill for a characteristic distance called ‘range’. In the example, directional variograms have been used to underline the spatial anisotropy of the phenomenon: less variability (or more continuity) in the horizontal direction (N45 and N90) rather than in the vertical direction (N0 and N135). This is directly linked to the spatial organisation of the dataset as lowest beta values are located in the upper part of the base map.

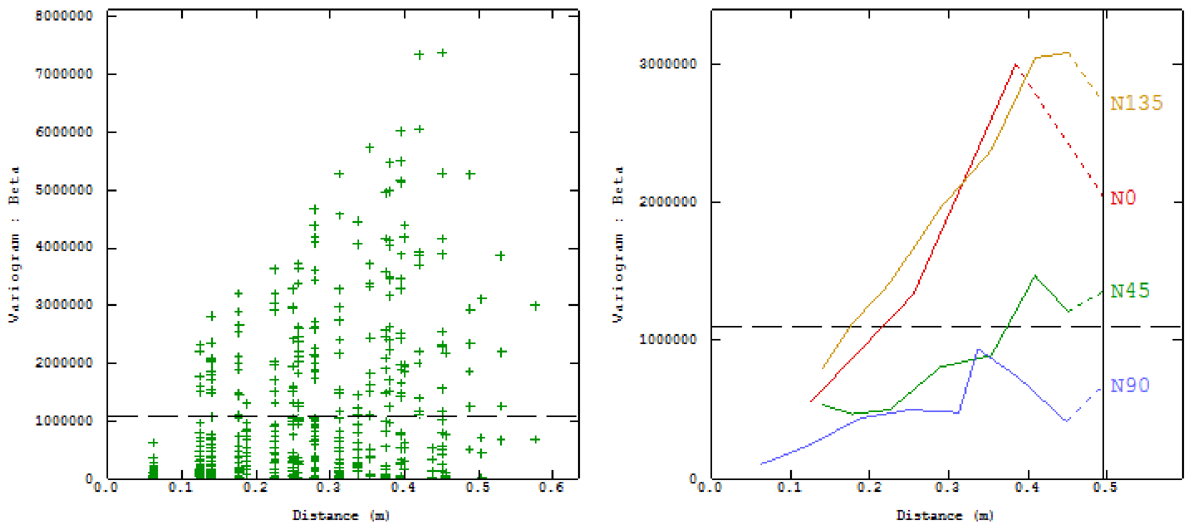

Figure 7: Variogram cloud (on the left) and directional variograms (on the right).

The variogram, which is based on data, allows interpretating the spatial continuity of the phenomenon. This spatial structure is crucial for the overall geostatistical approach. In the presented illustration, the anisotropy or spatial drift has to be taken into account.

Interpolation and uncertainty quantification

With input data and the spatial structure identified through the variogram, geostatistical techniques estimate the studied variable by a method called kriging (best linear unbiased estimator). This interpolation always includes a quantification of the associated uncertainty [Chiles and Delfiner, 1999].

More advanced and sophisticated geostatistical methods, such as conditional expectation or geostatistical simulations, can be used to provide other quantifications of uncertainties: risk of exceeding the threshold, for instance [Rivoirard, 1994]. These estimates are then powerful decision-making aids for the classification of surfaces and/or volumes prior to decontaminating works (based on different thresholds as well as considering the remediation support impact). For instance on Figure 8, with the same confidence interval on different estimates, the decision unit can be declared as clean if the probability of exceeding the threshold is below 50% and declared contaminated otherwise (probability above 50%). However the risk of misclassification is significant for 40% probability (false negative) and for 60% probability (false positive). According to other constraints (if the threshold is the release criteria or a segregation value between two rad waste categories, if the decision is about reuse, recycling or disposing of the material) the tolerated risk should not be the same. Similarly, with same estimates but with different confidence interval the risk changes according to the threshold level.

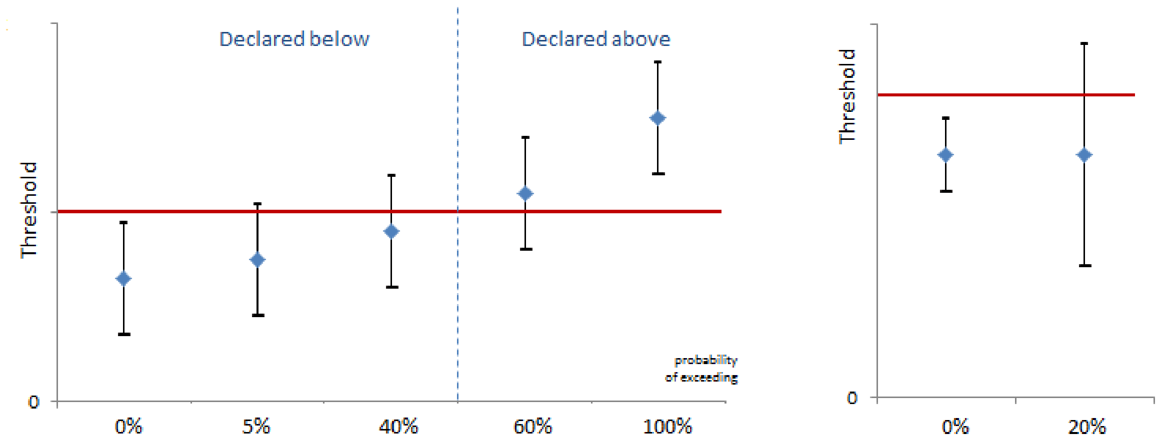

Figure 8: Probability of exceeding a threshold and misclassification risk with the same confidence interval (on the left) and with the same estimate (on the right).

In addition, multivariate geostatistics allows the combination of different kinds of information to improve estimates thanks to the spatial correlation between variables [Wackernagel, 1995]. Physical/historical data (such as matrices or information derived from incidents) and non-destructive measurements (for example dose rate or in situ gamma spectrometry) are advantageously integrated to improve the understanding and prediction of the main variable (results of laboratory analysis, for example) while reducing the estimation uncertainty.

Synthetic example on a linear metal structure

The piping network in the non-viable half culvert is not possible to access using the survey techniques described above for the viable whole culvert sections. In addition only limited data have been collected on the metal parts of the active culvert yet. Nevertheless, geostatistics is very useful to improve the radiological characterisation of contaminated metal objects and structures. A synthetic example is then created. It consists in a linear grid that can represent a metal pipe network in a building or in a culvert for example. Lengths and sampling resolutions are just given for information, not as recommendations.

Materials and data

To illustrate the added value of geostatistics for the radiological characterisation of metal materials to be recycled, a classical geostatistical study is performed on a synthetic example. The latter consists in a linear structure:

- Total length is 1,000 meters;
- Dose rates are measured at contact, regularly spaced every 50 cm;
- Samples are collected every 20 m for activity levels.

Contamination levels are preliminary obtained through a non-conditional simulation (using a 50m range cubic structure) on a 1D grid with a 10 cm mesh. Obtained values are then considered as the “real contamination”. Dose rates and activity levels are extracted from the simulated grid. A random variability is added for dose rate values (following a Gaussian distribution multiplied by the simulated value). This ensures a better representativeness of a real contamination as non-destructive measurements are generally associated with a limited variability due to background fluctuations, modelling assumptions (conversion coefficient), measuring conditions…

Comparison of results

The objectives are to estimate the contaminated length according to different thresholds from a global point of view (how much contaminated metal?) and from a local point of view (where is the contamination located?). As input values are known everywhere (from the initial simulation), a comparison is possible between estimation and true value.

A classification activity threshold (20) is considered. It can be a release criterion for example. Corresponding true values assume a total length of 494.6 m with contaminated pipe.

In absence of an advanced analysis, nearest neighbour interpolation technique is generally used. A total of 500 m is declared above the threshold of 20. However classification details presented in Figure 9 have to be explained:

- 453.6 m are correctly classified: both true value and estimated value above the threshold.
- 46.4 m are wrongly declared above the threshold whereas the true value is below (false positive, in blue).
- 41.0 m are wrongly declared below the threshold whereas the true value is above (false negative, in red).

That way, 4.6% of the metal materials are treated as contaminated ones. On the contrary, 4.1% are released whereas contaminated.

These results strongly impact the decommissioning project as regards cost, waste and impossibility to demonstrate clearance due to misclassification errors

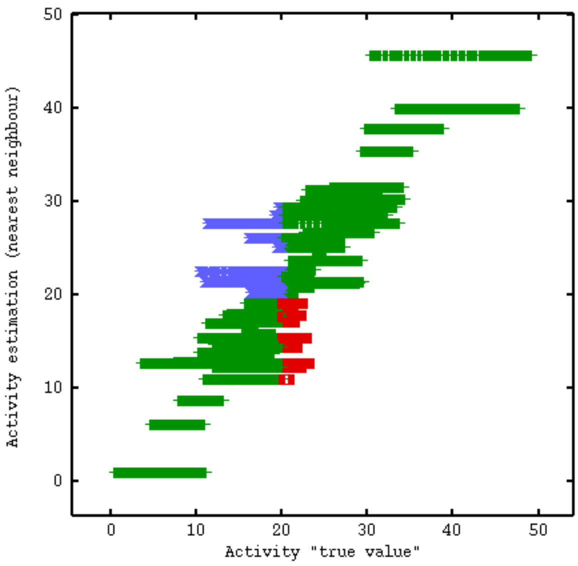

Figure 9: Comparison between deterministic estimation and true value and misclassification errors in blue (false positive) and in red (false negative).

Similar results are obtained with dose rate measures. Misclassification errors are less linked to the interpolation technique (as they are much more numerous) but strongly impacted by the measurement uncertainties.

In comparison with other estimation techniques, geostatistics provide a sound methodological framework to take the spatial structure into account as well as the decision support. In addition the added value is the uncertainty quantification for reliable decisions to be taken (classification according to a threshold or adding extra samples to reduce uncertainties).

On this synthetic example, geostatistics can be applied on the only activity levels (direct analysis, monovariate) and taking dose rate values into account (multivariate approach) in order to improve estimations and to reduce uncertainties. Kriging and co-kriging results are presented in Figure 10 for the first 200m. They are compared with the reality and the nearest neighbour interpolation technique.

Figure 10: Comparison of the different interpolation results for the first 200m.
Indication of the activity data points in black diamonds

Conditional simulations (direct and multivariate) are then computed and used to assess global (Figure 11) and punctual (Figure 12) risk curves.

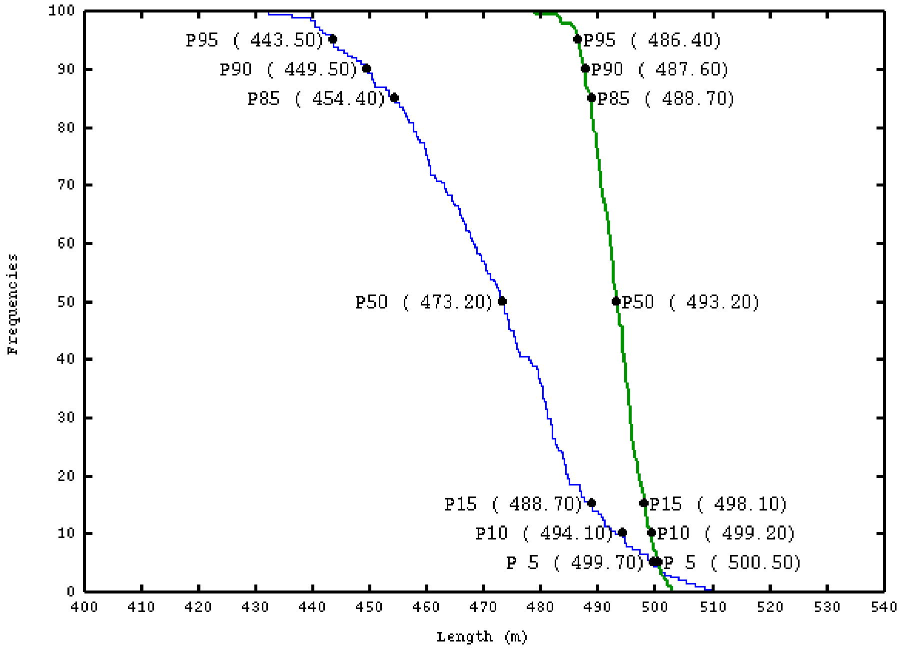

Figure11: Global estimation of pipe length above the threshold ignoring dose rate values (in blue) and including them (in green). Inverse cumulative representation.

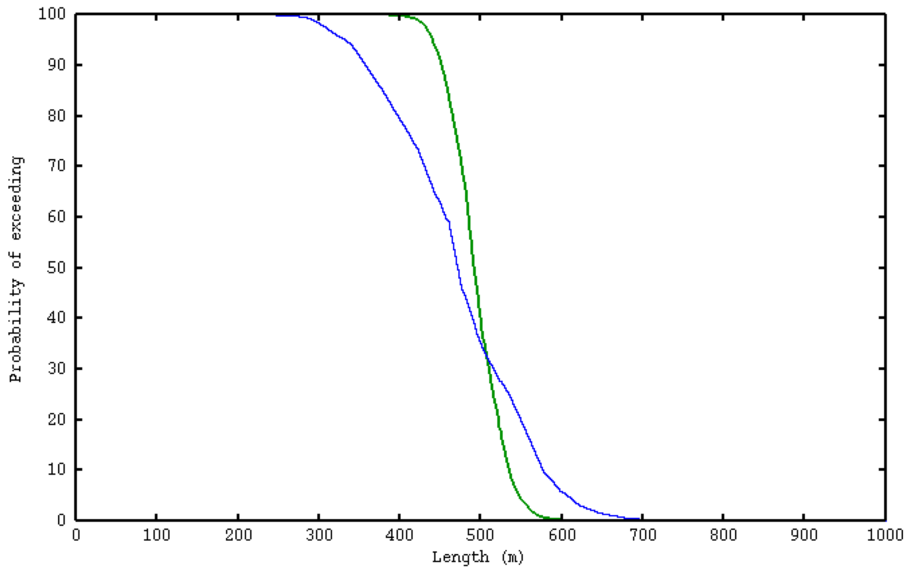

Figure 12: Punctual classification of pipe length ignoring dose rate values (in blue) and including them (in green). Inverse cumulative representation of the probability of exceeding the threshold.

First comment is that including dose rate values, even if they are blurred with uncertainty, improves the estimation both globally and punctually: risk curves are more vertical (higher slope), which means a reduction of confidence interval widths.

Then the true pipe length above the threshold (494.6m) is correctly estimated globally (answering to the first question: how much contaminated metal?) as the median value of the global risk curve is 493.2m and falling in the 90% confidence interval ranging from 486.4m to 500.5m. As for waste classification from a punctual point of view (answering to the second question: where?), the corresponding risk curve shows that the probability level can reasonably be considered around 10% (524.9m) as the slope is high before. Then a discussion can be engaged with the safety authority to reduce even more this risk down to 5% (547.4m) or 1% (559.2m). The supplementary quantity of materials is somehow quite reasonable as regards the total characterised pipe length to ensure a better confidence level.

Additional remarks

Sometimes geostatistics is not relevant as the volumes or the stakes are not important enough to justify a significant sampling effort and characterisation processing. But geostatistics demonstrated its added value for hundreds of nuclear sites and building under decommissioning and remediation (for soils and concrete structures) and also for metal materials such as heat exchangers, primary and secondary loops of nuclear reactors, nuke submarines, activated metal components of a graphite reactor, and so on.

Of course real datasets are generally more complex (different contamination thicknesses, different matrices, different fingerprints…). This reinforces the relevance of a comprehensive data analysis and the quantification of uncertainties. In addition, historical context and preliminary measurements (even gross counting) are valuable information that reduce uncertainties and allow optimizing sampling strategy.

Conclusion

A radiological survey has been performed in the active culvert at Studsvik, due to a planned decommissioning. However, parts of the metal piping network is difficult to access due to non-viable sections, and alternative survey methods are needed. The active culvert project gives the opportunity to elaborate a sound reflexion on the characterisation of contaminated metals. Even if limited data are available yet on the metal part of the piping network, the added value of relevant data processing for waste characterisation and categorisation of metals to be recycled is demonstrated. In addition sampling optimisation issue should be adequately tackled in coherence with the data processing.

Geostatistics proves to be a relevant data processing for a better characterisation of contaminated materials, metals in particular. That leads to a better segregation for clearance, recycling, reuse or waste minimisation.

As for the active culvert project, some metal parts of the piping network will be adequately characterised during the waste treatment. Other surfaces and components are characterised on-site for a better project management.

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