



## **GEOSTATISTICAL METHODS FOR RADIOLOGICAL EVALUATION AND RISK ANALYSIS OF CONTAMINATED PREMISES**

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### **ABSTRACT**

This paper presents an innovative geostatistical approach suitable for radiological evaluation in nuclear premises. By modelling the spatial continuity of activities, geostatistics provides sound methods to estimate and map radiological activities, together with their uncertainty. The main geostatistical principles are illustrated on real premises. Then, the paper investigates how it is possible to optimize the sampling strategy, to take historical information into account, and finally to quantify contaminated surface or volume uncertainties.

### **1 INTRODUCTION**

At the end of process equipment dismantling, the complete decontamination of nuclear facilities requires the radiological assessment of residual activity levels of building structures. As stated by the IAEA: “Segregation and characterization of contaminated materials are the key elements of waste minimization” [1]. From this point of view, the set up of an appropriate evaluation methodology is of prime importance. The radiological characterization of contaminated premises can be divided into three steps. First, the most exhaustive facility analysis provides historical and qualitative information. Then, a systematic (exhaustive or not) control of the radiation signal is performed by means of in situ measurement methods such as surface control device combined with in situ gamma spectrometry. Besides, in order to assess the contamination depth, samples can be collected at several locations within the premises and analysed. Combined with historical information and radiation maps, such data improve and reinforce the preliminary waste zoning.

The relevance of the geostatistical methodology relies on the presence of a spatial continuity for radiological contamination. In this case, geostatistics provides reliable methods for activity estimation, uncertainty quantification and risk analysis, which are essential decision-making tools for decommissioning and dismantling projects of nuclear installations.

Besides, the geostatistical framework provides answers to several key issues that generally occur during the clean-up preparation phase: How to optimise the investigation costs? How to

deal with data quality issues? How to consistently take into account auxiliary information such as historical inventory? How to integrate the remediation support into the modelling? How to quantify uncertainties in the remediation costs while computing contaminated volumes?

This geostatistical approach is currently applied to several former nuclear facilities of the CEA in France. The ATUE (enriched uranium workshops) premise, located in Cadarache, is a case in point [2][3]. Focusing on this premise, the paper presents the geostatistical methodology and its added value to: (i) optimise the sampling strategy, (ii) get a reliable mapping of the contaminated areas and (iii) estimate the corresponding waste volumes.

## 2 MATERIAL

For confidentiality reasons, all data presented in the paper have been multiplied by a constant value in order to conceal the real radiological levels. However, this modification does not change the spatial structure analysis; it only alters statistical results and colour scales.

### 2.1 Investigated Area

The “Atelier D” is one of the four workshops of the ATUE facility, Cadarache CEA Centre. For 30 years, it had been used for the recycling of uranium contained in different non irradiated scraps so as to transform it into nuclear purity products (mainly oxides) by liquid processes. The  $^{235}\text{U}$  enrichment was less than 10%. The historical analysis points out a few contamination incidents during the industrial exploitation, leaving a residual radiological contamination mainly located on the floor.

The workshop area is about 800 m<sup>2</sup>. The different processes were located in several rooms distributed along a central corridor through the building. The “Atelier D” is also composed of a basement and a floor. These parts are not presented here. Sloping roofs along the building are not part of the study area neither. All the process structures have already been dismantled and the building structures (mainly concrete) remain to be characterized and cleaned up.

### 2.2 Experimental Data

In 2008, an extensive non-intrusive measurement campaign has been carried out using surface detection systems and in situ gamma spectrometry. Surface measurements are realized with thin-layer plastic scintillation detectors for  $\alpha$  and  $\beta\gamma$ -radiation. Measurement values are proportional (confidentiality coefficient) to gross counting rates (cps). This paper is only based on  $\beta\gamma$ -radiation due to the presence of varnish, which makes the  $\alpha$ -radiation values inaccurate. Uranium is the only radioactive element within the building and is therefore characterized using the  $\beta\gamma$ -radiation of its decay products.

A regular 66-cm mesh leads to the realization of 1,617 measurement points on the floor (Figure 1), acquired in two weeks by two persons. The investigations carried on the workshop walls are not presented here.

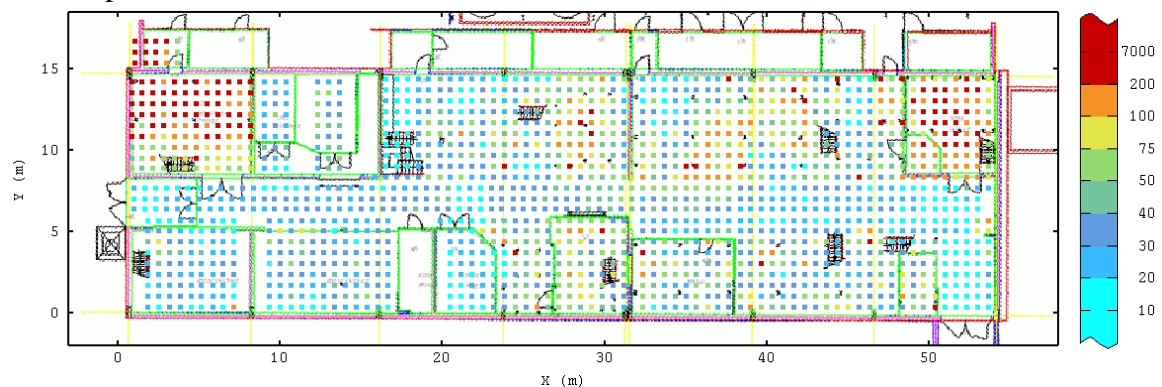


Figure 1:  $\beta\gamma$ -radiation (cps) with a 66 cm mesh in the "Atelier D" of ATUE facility.

In order to complete the radiological evaluation of the “Atelier D”, 1-cm depth concrete samples have been collected in 2009 from scabbling performed at 56 locations within the premises (Figure 2), determined on  $\beta\gamma$ -radiation maps. They have been analysed for uranium activity through  $^{235}\text{U}$  (expressed in Bq/g but here again, a confidentiality coefficient is used). The purpose of over-sampling in specific areas is to evaluate the variability of activities at small scale and assess the spatial representativity of the measured values.

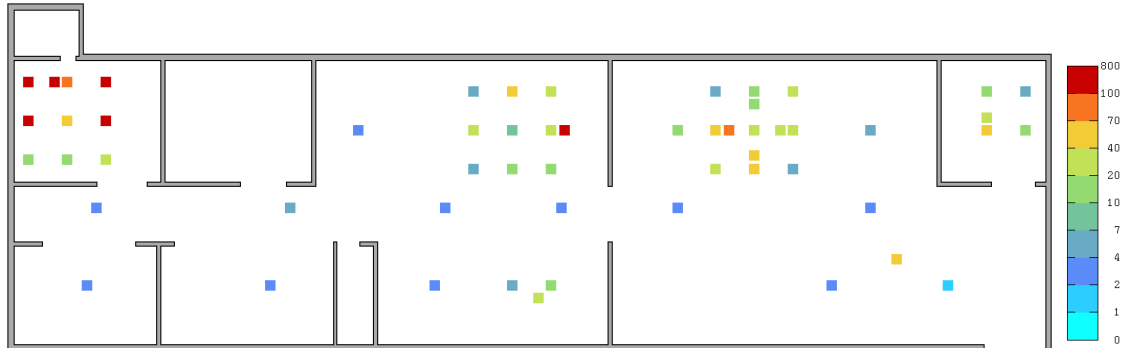


Figure 2: Uranium activity levels (Bq/g) at sampling points.

### 3 METHODOLOGY: GEOSTATISTICS

In order to present and illustrate the geostatistics framework, a classical geostatistical study is performed on the  $\beta\gamma$ -radiation values, from spatial structure analysis to mapping and uncertainty quantification.

#### 3.1 Spatial Variability and Variogram

The whole point of the geostatistical methodology is to take into account the spatial continuity of the phenomenon to predict it at unsampled locations, and quantify the prediction uncertainty. The characterization of this spatial continuity, or spatial variability, is an essential stage which is performed through the variographic analysis [4][5].

The experimental variogram is calculated by averaging, within classes of distance, the variability contribution of each couple of data; this contribution is usually quantified by the half squared difference of the measured values. Generally, for a structured phenomenon, the spatial variability increases with distance and tends to stabilize (“sill”) at a distance named “range”. Data separated by a distance larger than the range are no longer spatially correlated.

The raw data is classically transformed using a gaussian anamorphosis (intuitively, the raw histogram is deformed to become a Gaussian one). The resulting variogram is usually better structured, which facilitates the spatial structure determination (Figure 3).

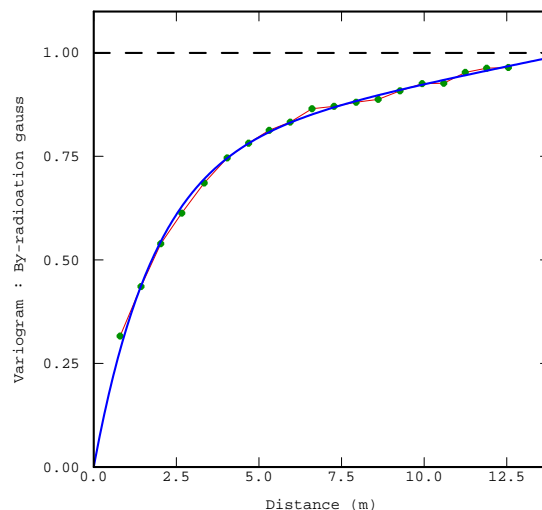


Figure 3: Experimental variogram points (green) and its fitted variogram model (blue).

The evolution of the spatial structure is clearly identified: the spatial variability quickly increases up to 5 m and then more gradually, which shows the existence of important changes of activity levels for a distance of 5 m.

The kriging (interpolation) procedure requires the model fitting of the experimental variogram. Indeed, for the following calculations, the spatial variability should be known whatever the distance and should integrate the a priori information on the phenomenon, which is not always illustrated by the measurements.

Of course, there is no unique model. Fitting is mainly based on the experimental variogram points and also on the fitting experience gained from similar radiological contamination datasets. The chosen variogram model (in blue on Figure 3) presents no discontinuity at the origin (no “nugget effect”) and is composed of an exponential structure with a practical range of 5.5 metres and a linear component.

### 3.2 Estimation by Kriging

Based on the measurement values and the previously fitted variogram model, the kriging procedure leads to an estimation of surface  $\beta\gamma$ -radiation. As for classical interpolations, kriging of the surface activity at a point  $x_0$ , denoted  $Z^*(x_0)$ , is a linear combination of the  $n$  known experimental values at measurement points:

$$Z^*(x_0) = \sum_{i=1}^n \lambda_i Z(x_i). \quad (1)$$

Kriging differs from other interpolators in the choice of the  $\lambda_i$  coefficients named “kriging weights”. They depend on: (i) the distances between the data and the point to be estimated (as for classical interpolators), (ii) the distances between the data (clusters...), and (iii) the spatial structure of the studied phenomenon (for example, very smooth or heterogeneous behaviour, anisotropy, etc., characterized by the variogram model).

The kriging weights are determined so as to ensure an unbiased estimation and the minimization of the estimation error variance  $\text{Var}[Z^* - Z]$ , which corresponds intuitively to the minimization of the error risk.

Kriging is the best linear unbiased estimator [6].

From a practical point of view, the estimation is carried out using the whole set of data (unique neighbourhood) or only the closest data points to the target point (moving neighbourhood). In our case, we use a moving neighbourhood of 30 points. The kriging results are presented on Figure 4.

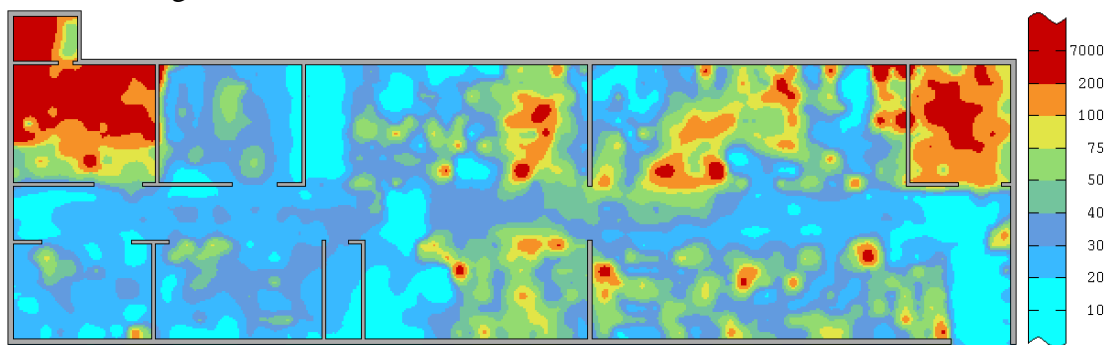


Figure 4: Kriged map of  $\beta\gamma$ -radiation (cps).

This kriged map points out several areas within the installation where the  $\beta\gamma$ -radiation is high. The conformity between historical analysis (contaminating work station, incidents...) and in situ measurements can be noticed. Moreover, contamination surface extensions are characterized through this cartography.

### 3.3 Kriging Standard Deviation

In comparison to classical interpolators, the added value of geostatistical estimation lies in the quantification of the related estimation uncertainty. This quantification is possible due to the spatial variability modelling.

Uncertainty is usually described by the kriging (error) standard deviation values: it takes minimal values close to data points, where the estimation confidence is high and it increases with the distance between the target point and the data points, as a function of the chosen variogram model.

Kriging standard deviation exclusively depends on the geometric configuration formed by the target point and the data points, and on the variogram model. This is its main advantage as it can be a priori calculated knowing the contamination spatial structure. This is also its limit because it does not consider the uncertainty variation related to activity levels.

Generally, the kriging standard deviation map is a good indicator of the estimation quality. In our case, the regular sampling pattern logically provides a regular kriging standard deviation map.

### 3.4 Uncertainty Quantification

Minimizing the error risk by construction, kriging smoothes extreme values, unlikely to be observed, and tends to bring them to the local contamination average. This is the kriging smoothing property: the contamination real variability is not reproduced through the interpolation step. Consequently, the kriging map cannot be directly used to estimate the probability to exceed a cutoff and the corresponding contaminated surface.

The Gaussian framework provides a more quantitative use of kriging standard deviation in terms of confidence interval for the activity levels.

The kriging procedure does not require any data statistical distribution hypothesis. But if the spatial distribution is Gaussian, so is the kriging error, then kriging results can be expressed in terms of confidence intervals (exactly in the case of simple kriging, in a first approximation otherwise). Since our raw variable has been transformed into a Gaussian one, confidence intervals defined for the Gaussian variable are then back-transformed in the scale of the original variable.

Such a distribution is applied to the  $\beta\gamma$ -radiation values. A risk analysis result (95% confidence interval around kriging estimations) is presented on Figure 5. The confidence interval takes minimal values close to data points and increases in the high-variability areas.

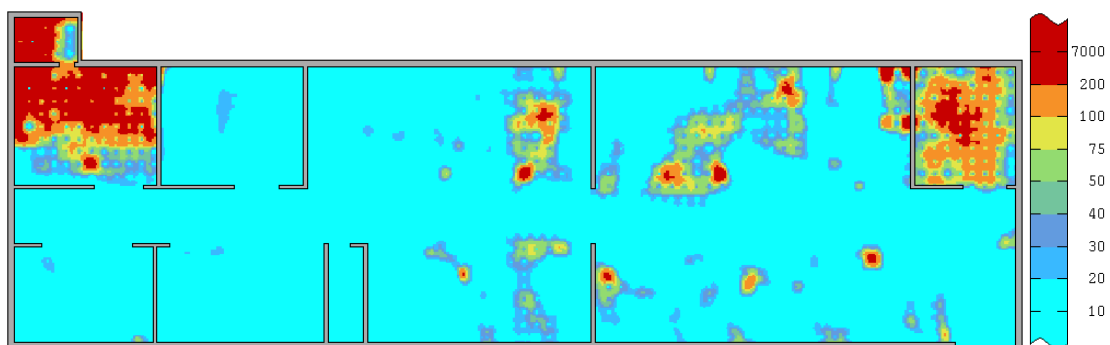


Figure 5: 95% confidence interval width around  $\beta\gamma$ -radiation kriging estimation (cps).

Likewise probability to be above a given cutoff can be calculated (with dismantling activity threshold for example). This kind of uncertainty maps may be used to position complementary investigations so as to reduce high uncertainty zones.

### 3.5 Geostatistical Simulations

Another way to quantify uncertainties lies in the use of conditional geostatistical simulations [4]. Each simulation corresponds to one possible scenario for the spatial distribution of the variable. All simulations are consistent with the variogram model and honour the available information (experimental values and statistical distribution).

Conditional simulations allow us to derive local estimates of non-linear quantities, such as quantile or probability maps, and to estimate global statistics like contaminated surfaces or volumes.

## 4 RADIOLOGICAL ASSESSMENT

### 4.1 Multivariate Variographic Analysis: Integration of Surface Measurements

So far, we only considered gross  $\beta\gamma$ -radiation measurements. Uranium activity levels are estimated through concrete samples, which are essential to perform a complete radiological characterization.

$\beta\gamma$ -radiation and uranium levels from concrete samples are both investigating the radiological contamination on the workshop floor. It is usual to realise the joint study of all data referring to the same phenomenon in order to take into account the link between data and improve the estimation process.  $\beta\gamma$ -radiation data is then integrated in order to improve the activity interpolation uranium activity levels.

Indeed, ignoring this radiation signal would lead to an important loss of information: uranium values are available at 56 points whereas radiation signal mesh is 0.66 m, which represents 1617 points (almost 30 times more). As for time and costs aspects, the comparison is clearly in favour of radiation measurements (quicker and cheaper).

The geostatistical methodology remains the same except that this is now a multivariate variographic analysis with two simple variograms for the two single variables and one cross-variogram which underlines the spatial behaviour of the correlation between the two variables (Figure 6).

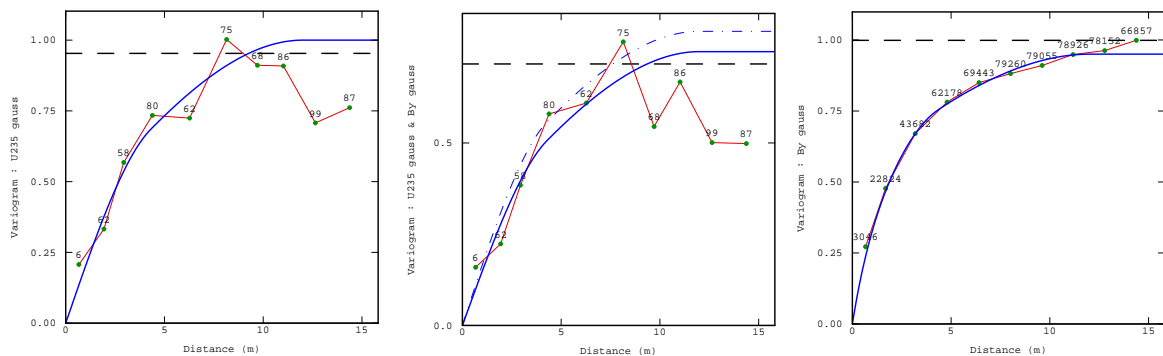


Figure 6: Simple variograms for uranium values (left) and  $\beta\gamma$ -radiation (right), cross-variogram (middle). Fitted variogram model in blue.

### 4.2 Uranium Activity Maps and Uncertainty

Estimation for uranium activity levels is then realised with and without  $\beta\gamma$ -radiation information. Results are presented on Figure 7 together with their uncertainty maps (95% confidence interval around the prediction).

The integration of the  $\beta\gamma$ -radiation data significantly improves the uranium activity level estimation, providing better defined contamination shapes. The impact of this auxiliary data on uncertainty maps is even more noticeable with a large reduction of the 95% confidence interval width, especially where only a few concrete samples (uranium activity levels) are available.

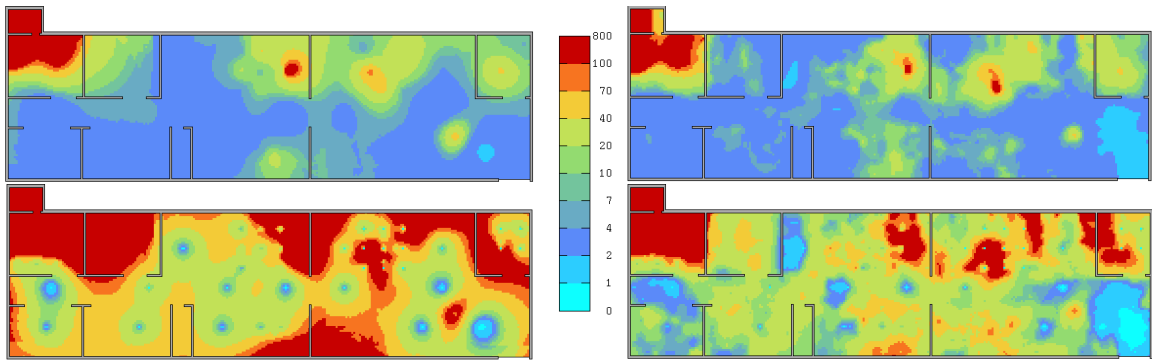


Figure 7: Activity maps (top) and their uncertainty maps (bottom) without the  $\beta\gamma$ -radiation data (left) and integrating this auxiliary data (right).

### 4.3 Decision Making-Aid Tools for Waste Segregation

Quantification of contaminated surfaces is realized by applying activity thresholds on conditional geostatistical simulations. The remediation support constraint is taken into account by the calculation in the different workstation areas, which are considered as an effective remediation support. For each cell of the grid, the probability to be greater or equal to a given cutoff is estimated by the proportion of simulated values that exceed the cutoff. Risk maps are presented on Figure 8 for two thresholds which can be interpreted, for example, as the risks to be above Very Low Level Waste criterion on the one hand and Low Level Waste criterion on the other hand. Logically, the higher the activity cutoff, the lower the probability to exceed this value.

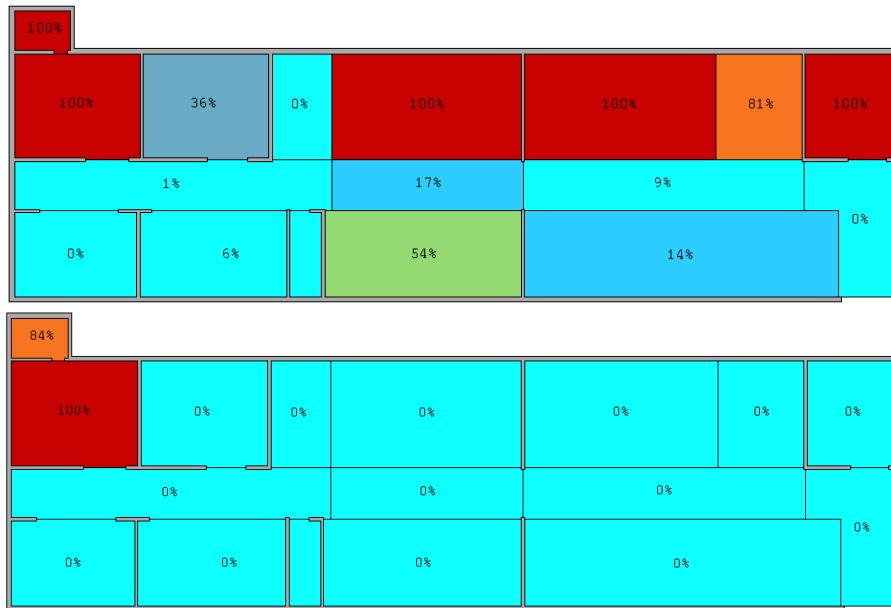


Figure 8: Risk maps for Very Low Level Wastes (top) and Low Level Wastes (bottom).

As for remediation costs, they naturally increase with the confidence level that is required. It is therefore possible to compute contaminated waste volumes depending on the probability to be above the activity threshold.

The quality and the number of data can strongly improve or deteriorate this kind of risk analysis. Decommissioning and dismantling projects are largely affected by the quality of this investigation phase, which has a significant impact on the risk level and the optimisation of the waste production.

## 5 CONCLUSIONS

This paper recalled the geostatistics principles and demonstrated how this methodology provides promising tools for the radiological evaluation of contaminated premises.

The relevance of such an approach relies on the presence of a spatial continuity for radiological contamination. In this case, geostatistics provides reliable activity estimates, uncertainty quantification and risk analysis, which are essential decision-making tools for decommissioning and dismantling projects of nuclear installations.

Besides, geostatistics can be employed so as to optimize the different sampling phases: integration of historical information, similarity of spatial structure with comparable contamination, relationship between different nuclear measurement devices (qualitative information of radiation maps combined with quantitative information of concrete sample activity levels).

## 6 ACKNOWLEDGMENTS

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## 7 REFERENCES

- [1] INTERNATIONAL ATOMIC ENERGY AGENCY, “Methods for the Minimization of Radioactive Waste from Decontamination and Decommissioning of Nuclear Facilities”, Technical Reports Series No. 401, IAEA, Vienna, 2001.
- [2] P. Lisbonne P., and Seisson D., “Enriched uranium workshops towards decommissioning Cadarache nuclear research centre”. Int. Conf. on Decommissioning Challenges: An Industrial Reality?, Avignon, France.
- [3] Desnoyers Y., Chilès J.-P., Jeannée N., Dubot D., Lamadie F., “Geostatistical sampling optimization and waste characterization of contaminated premises”. Int. Conf. Global 2009, Paris, France.
- [4] Chilès J.-P., Delfiner P., Geostatistics: Modeling Spatial Uncertainty, Wiley, New-York, 695p, 1999.
- [5] Matheron G., Estimating and Choosing - An Essay on Probability in Practice, Springer Berlin, 141p, 1989.
- [6] Isaaks E. H., and Srivastava R.M., An Introduction to Applied Geostatistics. Oxford University Press, 561p, 1989.
- [7] GEOVARIANCES 2009, Isatis Technical References, 9<sup>th</sup> Edition, Geovariances Fontainebleau, 222 pp, 2009.