**Proceedings of The ASME 2011 14th International Conference
on Environmental Remediation and Radioactive Waste Management
ICEM2011
September 25-29, 2011, Reims, France**

ICEM2011-59344

GEOSTATISTICAL METHODOLOGY FOR WASTE OPTIMIZATION OF CONTAMINATED PREMISES

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ABSTRACT

The presented methodological study illustrates a geostatistical approach suitable for radiological evaluation in nuclear premises. The waste characterization is mainly focused on floor concrete surfaces.

By modeling the spatial continuity of activities, geostatistics provide sound methods to estimate and map radiological activities, together with their uncertainty. The multivariate approach allows the integration of numerous surface radiation measurements in order to improve the estimation of activity levels from concrete samples. This way, a sequential and iterative investigation strategy proves to be relevant to fulfill the different evaluation objectives.

Waste characterization is performed on risk maps rather than on direct interpolation maps (due to bias of the selection on kriging results). The use of several estimation supports (punctual, 1 m², room) allows a relevant radiological waste categorization thanks to cost-benefit analysis according to the risk of exceeding a given activity threshold. Global results, mainly total activity, are similarly quantified to precociously lead the waste management for the dismantling and decommissioning project.

INTRODUCTION

For more than a century, the development of the French nuclear industry has led to the construction and exploitation of hundreds of facilities to produce nuclear fuel, burn it in experimental reactors or nuclear power plants, and eventually recycle it. Dozens of these facilities are now under decommissioning in France.

The complete decontamination of nuclear facilities requires the radiological assessment of residual activity levels of building structures. As stated by the International Atomic Energy Agency [1]: “Segregation and characterization of contaminated materials are the key elements of waste minimization.”

In this framework, the relevance of the geostatistical methodology relies on the presence of a spatial continuity (for radiological contamination), characterized through the variographic analysis. Geostatistics then provides reliable methods for activity estimation, uncertainty quantification and risk analysis [2], which are essential decision-making tools for decommissioning and dismantling projects of nuclear installations. For less than a decade, geostatistics has successfully been used for the radiological evaluation of contaminated sites [3] but nothing exists, to the best of the authors’ knowledge and except previous work from the authors [4], for its application to nuclear facilities.

Besides, the geostatistical framework provides answers to several key issues that generally occur during the decontamination preparation stage: How to optimize 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 modeling? 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 CEA Center, is a case in point. Focusing on this premise, the presentation deals with geostatistical methodology and its added value to get a reliable mapping of the contaminated areas and estimate the corresponding waste surfaces or volumes.

METHODOLOGY AND MATERIALS

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, nor the methodological approach.

Evaluation Methodology for the Categorization of Radiological Waste

Decommissioning and dismantling projects are largely affected by the quality of the investigation stage, which has significant impacts on the estimated risk levels and waste segregation optimization. The quality and the number of data for the characterization can strongly improve or deteriorate the risk analyses, affecting global remediation costs [4].

The proposed methodology for the radiological characterization of contaminated premises is divided into three steps according to the three available levels of information:

1. First, the most exhaustive facility analysis provides historical and qualitative information: functional analysis, incidents, isotopy;
2. 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;
3. Finally, in order to assess the contamination depth, samples are collected at several locations within the premises and analyzed.

Combined with historical information and radiation maps, the analysis of activity levels improves and reinforces the preliminary waste zoning required by the French Nuclear Safety Authority [5].

Investigated Area

"Atelier D" is one of the four workshops of the ATUE facility, Cadarache CEA Centre [6]. For 30 years, it was 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}U enrichment was less than 10 %.

The workshop area is about 800 m². The different processes were located in several workstations distributed along a central corridor. All the process equipments have already been dismantled whilst the building structures (mainly concrete) remain to be characterized and cleaned up.

The functional analysis provides a well-documented workshop organization (processes, liquid flows…) and distinguishes two types of processes according to the nature of the uranium product: liquid phase or solid state. The historical analysis points out a few contamination incidents during the industrial exploitation that left a residual radiological contamination essentially located on the floor.

Experimental Data

In 2008, an extensive non-intrusive measurement campaign was carried out using surface detection systems and in situ gamma spectrometry. This campaign corresponds with the second step of the characterization methodology, which is a key element for the analysis of the contamination spatial extension and also for the optimization of destructive investigations (both number and location).

Surface measurements (10 to 15 seconds) are realized with thin-layer plastic scintillation detectors for α and βγ-radiation. Measurement values are proportional to gross counting rates (cps). The paper focuses on βγ-radiation as the presence of varnish makes the α-radiation values inaccurate. Uranium is the only radioactive element within the building and is therefore characterized using the βγ-radiation of its decay products.

A regular 66 cm mesh leads to the realization of 1,617 measurement points on the floor (Fig. 1). The investigations carried out on the workshop walls and on specific areas (judgmental approach) are not presented here.

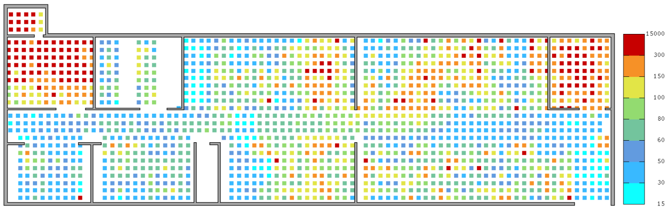

Fig. 1. βγ-radiation measurements (cps) with a 66 cm mesh in the "Atelier D" of ATUE facility.

The statistical distribution (Fig. 2) of βγ-radiation shows a strong dissymmetry with a few very high values and a lot of values around the background noise where there is no contamination. The distribution is presented using a log scale in order to better capture the range of radiation measurements.

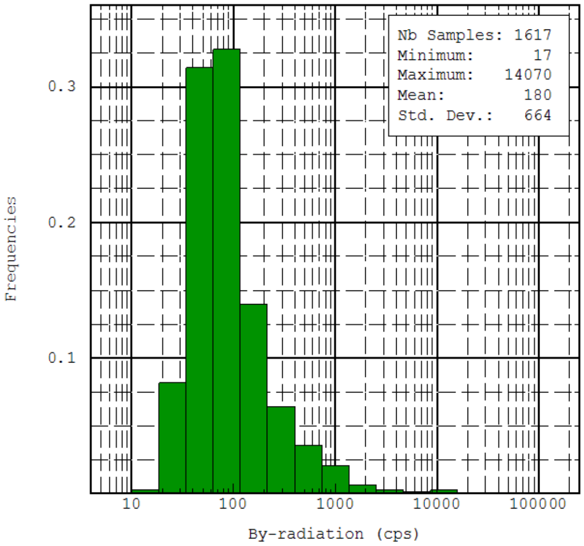

Fig. 2. Histogram of the raw βγ-radiation values using log scale. Classical statistics.

In order to complete the radiological evaluation of the workshop, 1-cm depth concrete samples were collected in 2009 from scabbling performed at 56 locations within the premises, chosen from βγ-radiation maps. These samples have been analyzed for uranium activity through ^{235}U (expressed in Bq/g with a confidentiality coefficient). The purpose of the over-sampling in specific areas is to evaluate the variability of uranium activities at small scale and assess the spatial representativeness of the measured values.

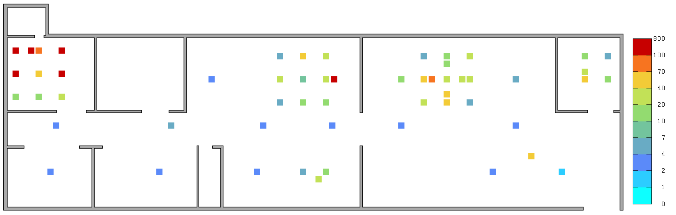

Fig. 3. Uranium activity levels (Bq/g) at sample points.

RADIOLOGICAL MAPPING USING GEOSTATISTICS

It is usual to perform the joint study of all data referring to the same phenomenon in order to take into account the link between data and ameliorate the estimation process. As βγ-radiation from surface measurements and uranium levels from concrete samples are both investigating the radiological contamination on the workshop floor, surface radiation data is then integrated in order to improve the interpolation of uranium activity levels for the first centimeter of floor concrete.

Indeed, ignoring this radiation signal would lead to an important loss of information: uranium values are only 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 favor of radiation measurements (non destructive, quicker and cheaper).

Multivariate Variographic Analysis

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 [7].

The experimental variogram *γ(h)* is calculated by averaging, within classes of distance *h*, the variability contribution of each couple of data; this contribution is usually quantified by the half squared difference of the measured values:

$$\gamma(h) = \frac{1}{2} E\left[Z(x) - Z(x+h)\right]^2 \qquad \text{(Eq. 1)}$$

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.

Due to the strong dissymmetry of the statistical distribution (as presented in Fig. 2 for βγ-radiation), 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 identification. In addition, this preliminary transformation gives access to more sophisticated results (non-linear quantities).

The kriging (interpolation) procedure requires the 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.

In the multivariate case, 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 behavior of the correlation between the two Gaussian transformed variables (Fig. 4).

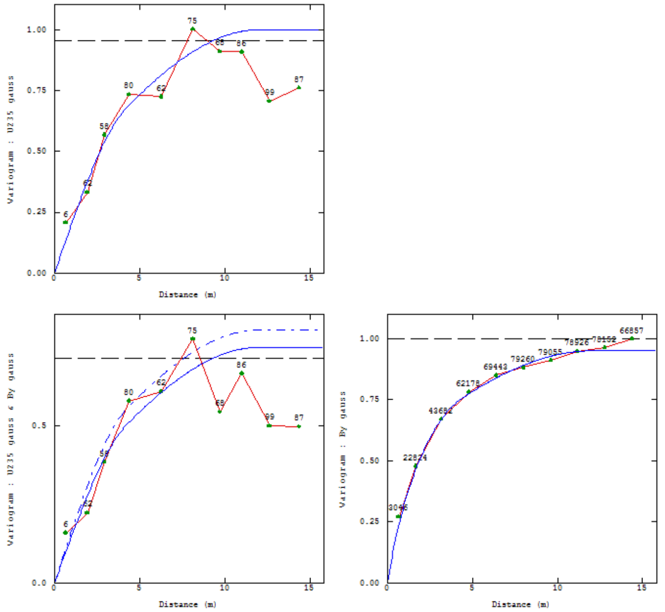

Fig. 4. Simple variograms for uranium values (top left) and βγ-radiation (bottom right), cross-variogram (bottom left). Fitted variogram model in blue.

The variogram fitting in the multivariate case should ensure theoretical properties (positive variances…). In order to set a coherent variogram modelling, the linear model of coregionalization is commonly used: it consists in a linear combination of the same basic structures, notably respecting the Cauchy-Schwartz inequality. Details on the geostatistical multivariate theory can easily be found in the literature (e.g., [8]).

Uranium Activity Maps and Uncertainty

Cokriging is the multivariate version of kriging, as a linear combination of all available data. In our case, uranium activity level is considered as the principal variable, known at 56 sample points, and βγ-radiation level is the auxiliary variable, more exhaustively available due to the 66 cm regular mech. Then calculations ensure the best linear unbiased estimation by minimizing the error variance.

Estimation for uranium activity levels is then realized with and without βγ-radiation information (Fig. 5 versus Fig. 6). Interpolation results are presented together with their uncertainty maps. Indeed, the added value of geostatistical estimation lies in the quantification of the related uncertainty, which is possible due to the spatial variability modeling. The Gaussian framework provides a more quantitative use of the cokriging standard deviation in terms of confidence interval for the activity levels. In the present case the 95% confidence interval around the prediction is chosen as the uncertainty indicator on the different maps.

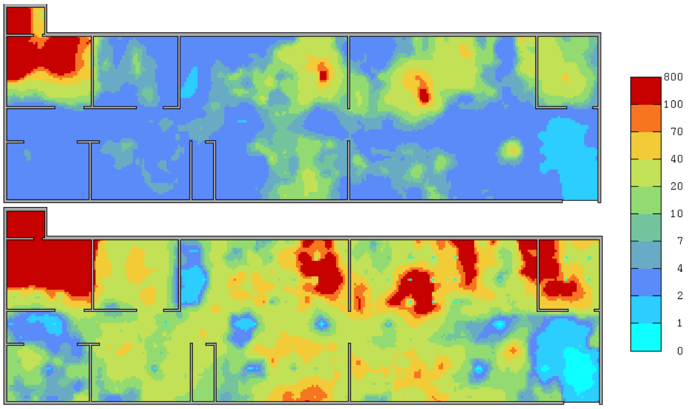

Fig. 5. Uranium activity map (top) and related uncertainty (bottom) integrating βγ-radiation data (cokriging).

The integration of the βγ-radiation data significantly improves the uranium activity level estimation, providing better defined contamination shapes (smooth and round shapes in the kriging case). 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.

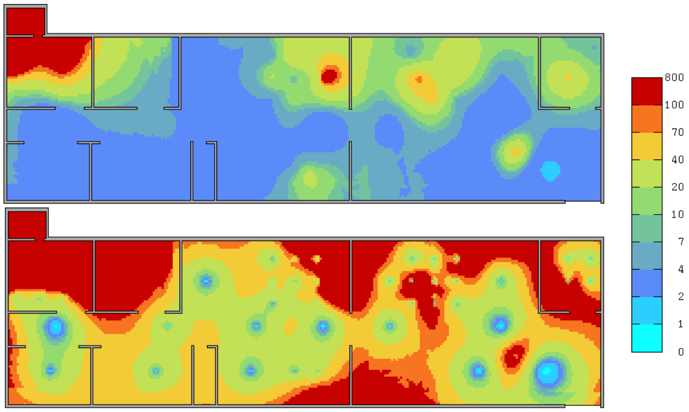

Fig. 6. Uranium activity map (top) and related uncertainty (bottom) without βγ-radiation data (kriging).

Uncertainty quantification allows an intelligent interpretation of the estimation results: areas with significant contamination, with high variability (transition zones between activity levels), or under-sampled areas (due to extrapolation) are easily indentified this way. Given the evaluation objective, it then facilitates the positioning of additional measurements or samples to reduce uncertainties.

As a consequence, the geostatistical multivariate processing reinforces the proposed sequential evaluation methodology, allowing a global sampling rationalization (cost and delay) between historical knowledge, in situ measurements and destructive samples.

The application of the geostatistical methodology on this dataset was described in details in [4] with a particular emphasis on sampling optimization according to spatial structure and historical information (liquid phase or solid state).

ESTIMATION SUPPORT AND WASTE SEGREGATION

Waste categorization is one key element for radiological waste management during nuclear facility dismantling. The geostatistical framework is particularly relevant to waste categorization through risk analyses and well-suited to take the different decontamination supports into account.

Geostatistical Simulations for Risk Analysis

In our case, cokriging is designed for the punctual estimation of uranium activity levels. The point is now to perform risk analysis according to a given threshold and considering a larger estimation support.

By construction, (co-)kriging smoothes the variability. Risk analyses based on the interpolation results would lead to inaccurate estimations by ignoring the related uncertainty.

Geostatistical simulations precisely focus the spatial variability reproduction (Fig. 7). Each simulation corresponds to one possible scenario for the spatial distribution of the variable. All simulations are consistent with the variogram model and honor the available information (experimental values and statistical distribution).

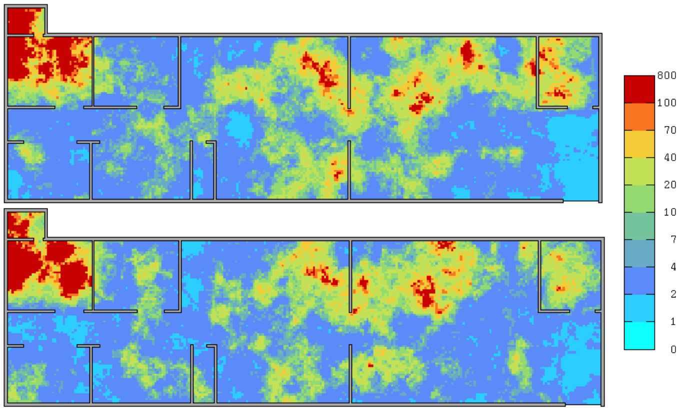

Fig. 7. Two examples of conditional cosimulation of uranium activity levels integrating βγ-radiation data.

Conditional simulations give access to local estimates of non-linear quantities, such as quantiles or probability maps. They are also designed to estimate more global statistics like the amount of contaminated surfaces or volumes and the total amount of activity (source term).

Global estimation of activity accumulation

Simulations provide a statistical distribution of the source term (accumulation curve). Here again, the added value of the multivariate approach through the integration of the βγ-radiation data leads to a significant reduction of the uncertainty related to the estimated quantity. As visible in Fig. 8 as an inverse cumulative histogram of total activity, the use of the auxiliary information leads to a 20% decrease of the median value for the source term and a reduction by a factor 3 of the 90% confidence interval width.

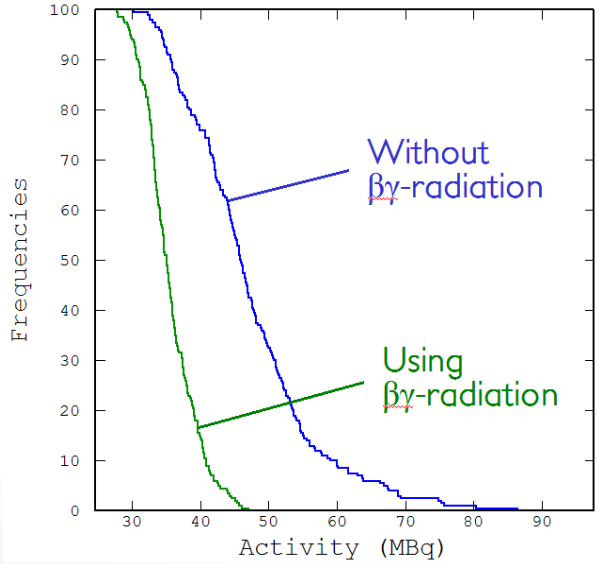

Fig. 8. Distribution of the global amount of uranium activity taking into account the βγ-radiation data (in green) or not (in blue).

This global result allows the quantification of radiological activity of all wastes to be produced during the decontamination and dismantling of the facility.

Waste Segregation and Estimation Support

Quantification of contaminated surfaces is performed by applying uranium activity thresholds on a large number of these conditional geostatistical cosimulations between uranium activity levels of sparsely collected concrete samples (principal variable) and radiation levels of the more numerous surface measurements (auxiliary variable).

The remediation support constraint is taken into account by considering the different workstation areas as effective remediation supports (as regards decontamination techniques). The central corridor is split into several parts to distinguish areas. The same can be done within each workstation if relevant.

Using the punctual simulations, the probability of exceeding a given activity threshold within each area is computed leading to an effective cost-benefit analysis. These calculations thus address the tricky change-of-support problem in a simple way. Results are compared for punctual support, 1 m²-support and workstation support for a radiological threshold of 10 Bq/g for uranium, see Fig. 9. Similar results may be obtained for various activity levels in relation with radiological waste thresholds.

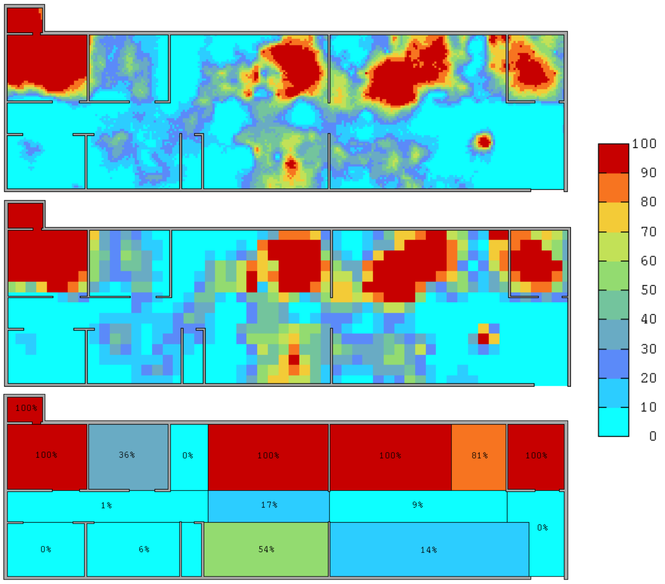

Fig. 9. Probability map of exceeding a given uranium activity level for each workstation.

Results are consistent and the support effect "averages" the punctual probability on larger areas. This way, punctual results are employed to identify hot spots that may be removed first due to radioprotection considerations. Indeed a single hot spot can be the cause of a non negligible probability of exceeding a given activity threshold at the workstation-support scale. This is the case for the workstation area with a 14% probability (Fig. 9), with a significant reduction of the risk (only in the right part because 20% to 60% risk areas are numerous in the left part).

In addition, 1 m²- or workstation-support maps are important decision tools to estimate the waste volumes to be produced. Considering a given activity threshold (10 Bq/g for uranium in our case), cost-benefit analyses are performed by comparing the risk threshold and the corresponding waste surfaces or volumes, see Fig. 10. Similar risk analyses may be used for other uranium activity thresholds in relation with the different radiological waste categories.

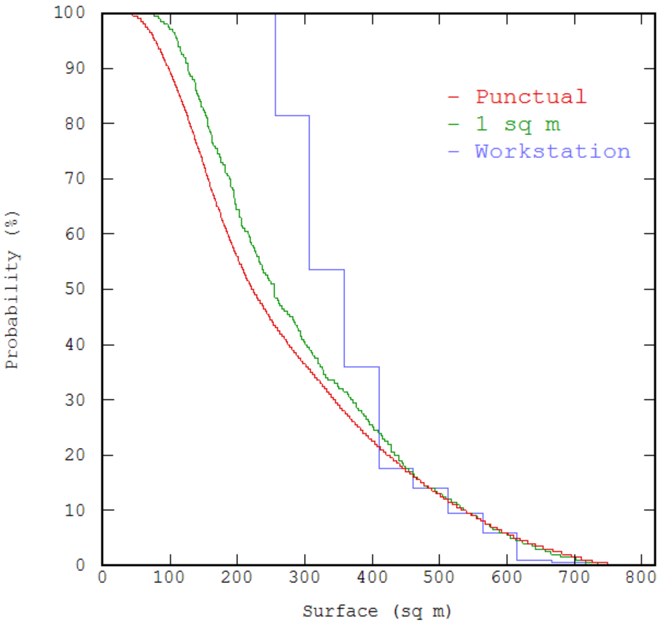

Fig. 10. Surface segregation for 10 Bq/g according to the tolerated risk and the calculation support.

As for remediation costs, they naturally increase with the required confidence level: the lower the risk, the larger the corresponding surface. The risk to be considered mainly depends on the activity threshold (between different radiological waste categories). The support effect has a strong impact on high probability (low risk) results with the dilution effect when uncontaminated areas (in comparison with the threshold) are mixed with contaminated ones within a workstation area.

The quality and the number of data can strongly improve or deteriorate this kind of risk analysis. As a consequence, decommissioning and dismantling projects are largely affected by the quality of the investigation stage, which has significant impacts on the estimated risk levels and waste segregation optimization.

CONCLUSIONS

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

The relevance of this 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.

Waste characterization is then performed taking all relevant information into account: historical knowledge, surface measurements and samples. Thanks to the multivariate processing, the different investigation stages can be rationalized as regards quantity and positioning.

Waste characterization is finally obtained through the analysis of probability maps of exceeding activity levels. The estimation support must be taken into account to discriminate punctual issues, such as hot spot identification, and waste production issues on larger areas. The main goal of this data processing remains an easier radiological waste management and the best waste categorization with acceptable investigation costs.

Ongoing research deals with the implementation of the geostatistical methodology on nuclear systems and equipments and on former radiological waste storages.

ACKNOWLEDGMENTS

This work is supported by GEOVARIANCES and the Geostatistics Group of MINES ParisTech. Besides, an industrial partnership with CEA partially funded the work and gave the access to nuclear facilities.

All geostatistical calculations and graphics are performed using ISATIS software [9].

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