CHARACTERIZATION OF A DEEP RADIOLOGICAL CONTAMINATION: INTEGRATION OF GEOSTATISTICAL PROCESSING AND HISTORICAL DATA

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KEY WORDS: Radiological contamination, Geostatistics, Historical data, Iterative campaigns.

ABSTRACT

The problem of site characterization is quite complex, especially for deep radiological contamination. This paper illustrates the added value of geostatistical processing on a real application case dealing with soils around facilities partially dismantled at the end of the 1950s in Fontenay-aux-Roses CEA Center (France).

12 years ago, a first exploratory drillhole campaign confirmed the presence of a deep radiological contamination (more than 4 m deep). More recently, 8 additional drillholes failed to delineate the contamination extension.

The integration of the former topography and other geological data led to the realization of 10 additional drillholes. This final stage significantly improved the characterization of the radiological contamination, which impacted the remediation project and the initially estimated volumes.

INTRODUCTION

Emerging in the early 80's, decommissioning is more than ever a major issue since hundreds of sites and facilities worldwide will end their operations over the next decades. Decontamination and remediation projects are all the more sensitive since they could last several years and turn out to be highly costly if not well-prepared. The key lies in an adequate contamination knowledge which helps to manage the remediation works and optimize the radiological waste production.

The commonly applied methodologies base their decision-process on more or less complex statistical analyses aiming at validating the final radiological state after the waste removal work (assuming the spatial randomness of values). These guides and norms are suited for demonstrating compliance with a dose- or risk-based regulation (French Nuclear Safety Authority 2010, US EPA 2000). However these techniques ignore the spatial behavior of the contamination and the importance of sampling strategy for the initial characterization, prior to the remediation works. The lack of representation and data processing tools may lead to inefficient radiological characterizations, which always maximize the amount of contaminated soils or concrete volumes.

To solve these issues, the French Atomic Energy Authority (CEA) has developed a methodology over the last 10 years with Geovariances' partnership to fulfill the radwaste categorization (Dubot et al. 2010). This methodology consists in an ordered sequence of evaluation actions starting with historical and functional analyses, in-situ characterization using non-intrusive measurement techniques if relevant, validation of contamination activity levels and depths with drillholes and laboratory analyses. In this framework, geostatistics gives more value to the collected data and allows mapping the contamination at each step of the sequence to finally get a robust and reliable characterization of contaminated areas (Goovaerts 1997). It also provides an efficient way out for sampling network optimization.

The paper introduces the geostatistical methodology and illustrates its added value on a real application case dealing with soils around facilities partially dismantled at the end of the 1950s.

INVESTIGATIONS

Historical Context

The CEA center of Fontenay-aux-Roses was created just after World War II initially to house nuclear research and development, both civilian and military, in France.

The first generation of nuclear facilities (ZOE, the first French nuclear reactor, uranium enrichment plant, plutonium recycling plant, laboratories…) had initially been implanted on the former military fortification of Chatillon, 5 km south of Paris. A large part of these facilities were dismantled in the 1950s and 1960s to build the second generation of nuclear facilities (Triton reactor, radiochemistry laboratory…) nowadays dismantled or under decommissioning. A third generation of buildings is now dedicated to biomedical research.

Chronology of the Investigation Campaigns

The complete dataset was collected over more than a decade by several drillhole campaigns (see Fig. 1):

- 12 years ago, a first exploratory drillhole campaign confirmed the presence of a deep radiological contamination (more than 4 m deep) during a judgmental survey of the site grounds (4 red points in the base map). The area was then classified as having a radiological contamination to be further investigated.
- In 2007, 8 additional drillholes (green points) intended to delineate the contaminated volumes around the initial and contaminated drillhole (as an horseshoe or half a circle), but failed.
- In 2009, 4 drillholes (dark points) were carried out along a quarter of a second circle centered on the initial contaminated point.
- According to the advanced analysis of the contamination (see corresponding section hereafter), 6 last drillholes (blue points) were performed in 2010. One of them was located close to the initial and contaminated drillhole; one was located within a nearby building.

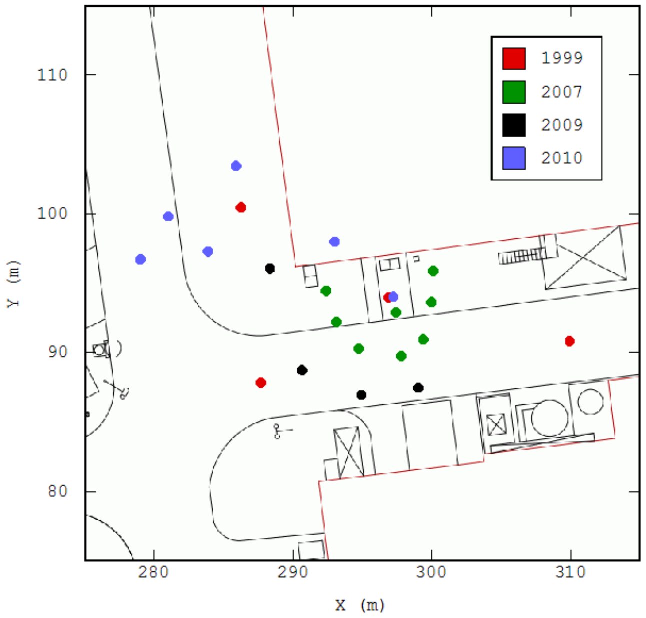

Fig. 1. Base map of the several drillhole campaigns.

Drillhole depths vary from 6 m to 15 m. Samples were collected with different lengths according to the geological information (from 5 cm to 50 cm). Gamma scanning was performed on cores from 2007 drilling campaigns on.

Samples were analyzed for ^{137}Cs and for other nuclides like ^{90}Sr, $^{239+240}$Pu, etc. Results are only presented for ^{137}Cs as this nuclide stands for a good tracer of the radiological contamination.

Activity values of the very first drillhole (1999) are unavailable as contamination was not expected at that level: the core was partly fouled due to cross-contamination from the water drilling technique. As a consequence, dry sonic drilling technique was preferred for the next drillholes.

For confidentiality reasons, all color scales and values presented in the paper have been masked to conceal the real radiological levels. However, this modification does not change the following geostatistical analyses, nor the methodological approach.

RESULTS AND DISCUSSION

Reminders of geostatistics and its application to soil pollution may be found in Chilès and Delfiner (2012), Goovaerts (1997) or Geosipol (2005). Desnoyers *et al.*(2009) intended an application of the geostatistical framework for the radiological characterization of contaminated premises and Desnoyers (2010) presents the geostatistical methodology applied to radiological contamination.

Initial Characterization of the Contamination

The first spatial characterization of the contamination is only based on the 2007 drillholes. Sample analyses show a strong dissymmetry of the activity value: a lot of measurements below the detection limit and a few high activity levels (Fig. 2).

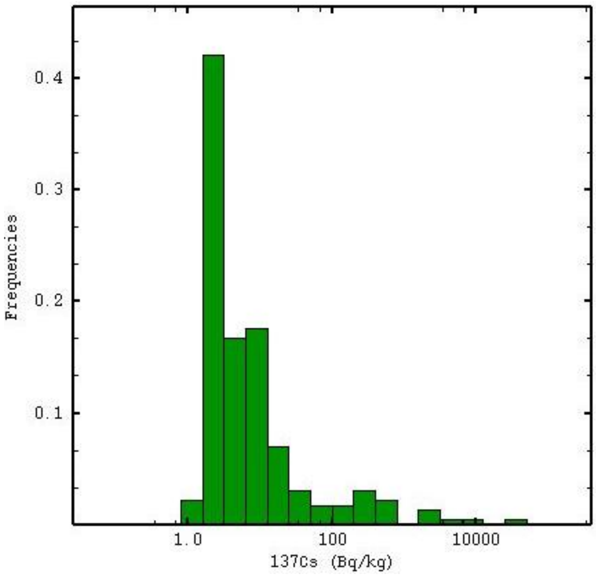

Fig. 2. Histogram of ^{137}Cs values in logarithmic scale.

At first, geostatistics is not directly applied on the raw data but on its log transformation. This is one easy and common technique applied to take the observed dissymmetry into account (but not compulsory). The variographic analysis does not show a convincing spatial evolution of the variability due to the small number of samples at a given depth resulting from the spatial configuration of the drillholes (7 points over half a circle plus its center).

However, a spatial interpolation is performed assuming an isotropic spatial structure (with no nugget effect). Kriging results clearly identify a contamination between 4 and 6 m in depth (Fig. 3), confirming the detected contamination on the very first drillhole in 1999.

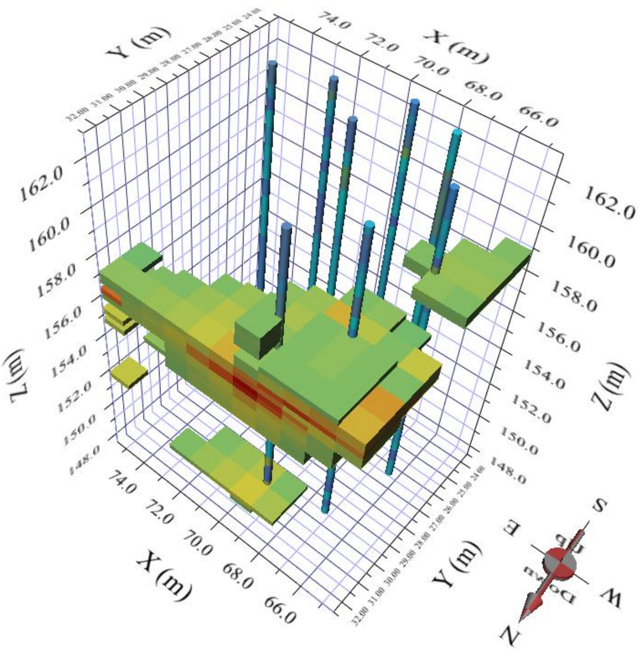

Fig. 3. Three-dimensional representation of the contamination with the first geostatistical analysis. Selection of high activity estimates (kriging).

Nevertheless, this preliminary study faced a lot of uncertainties for the characterization of the contamination:

- The spatial structure of the contamination is modeled under various hypotheses due to the lack of information in the horizontal plane;
- The contamination is not bounded yet in the North and West directions;
- The origin of the contamination in depth may not be linked with the current surrounding buildings and is still unknown.

Advanced Characterization of the Contamination

For all these reasons, additional drillholes were performed in 2009 on the North-Western part of the investigated area. Surprisingly they just identified a very thin contaminated layer precisely at a depth of 8 meters (2 meters below the contamination described in the initial characterization).

Until 2007, the observed activity values were interpreted as a deep contamination under a road, between two buildings of the second generation. The newly identified and deeper contaminated area drew our attention to the fact that the contamination may be connected with the previous site configuration, which was the military fortification with the first generation of nuclear facilities. Thus the base map is updated with the former site configuration (see Fig. 4): contaminated samples were in fact collected in the continuation of pools collecting contaminated water from the former plutonium recycling plant, along a bank and in the former moat. See also Fig. 7 for a transverse section of the moat (with the former topography and the mapping results).

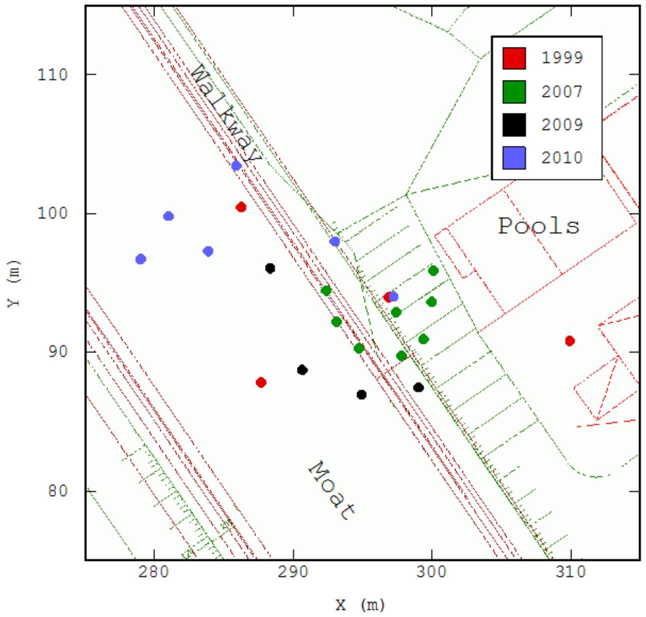

Fig. 4. Base map of the drillhole campaigns with the former map of the site.

At that stage, the last 6 drillholes (blue points in the base map) were performed in 2010: two of them located in the walkway, the other ones in the moat to delineate the extension of the contamination at 8m depth.

The geostatistical study has then been updated. Due to the strong dissymmetry of the statistical distribution of activity levels (as described previously), the raw data is now transformed using a Gaussian anamorphosis (intuitively, the raw histogram is deformed to become a Gaussian one). In addition to a better spatial structure identification with the resulting variogram (see Fig. 5), this classical transformation in geostatistics gives access to more sophisticated results (non-linear quantities such as probability of exceeding a threshold).

Due to the fitting constraints on the directional variograms (anisotropy of a single exponential structure), the spatial variability is slightly overestimated in the vertical direction after 1.5 m. However, this has little (and conservative) impact on estimation results due to the number of samples along the drillhole usually distant from less than 1.5 m.

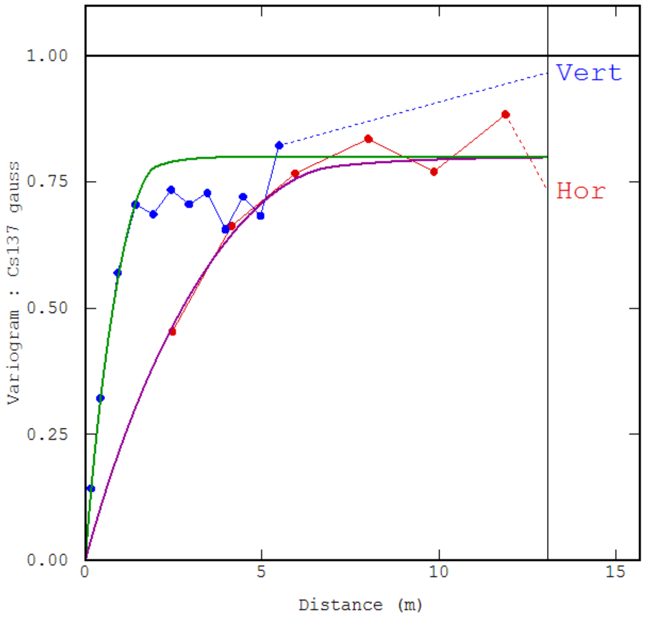

Fig. 5. Experimental variograms and corresponding models, in the horizontal plane (Hor) and in the vertical direction (Vert).

Contamination is then characterized through geostatistical simulations in order to estimate probability of exceeding radiological thresholds within the multi-Gaussian framework (thanks to the Gaussian anamorphosis).

In Fig. 6, the 3D representation of the contamination is given for an activity threshold, 10 times higher than the highest detection limit in ^{137}Cs. Only 30% and above probabilities of exceeding the threshold are presented.

The former topography is interpolated using spline model kriging after the digitalization of the elevation information on the map. Faults are used to improve the modification of the moat walls. Former buildings are also presented.

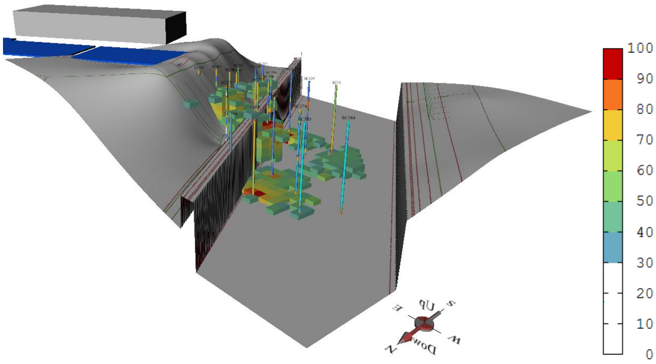

Fig. 6. Three-dimensional representation of the contamination in the former site context (topography, pools and moat). Only probabilities > 30% are visible.

The cross-section of the site, presented in Fig. 7, perpendicular to the moat, underlines several contaminated areas: a thin layer at the bottom of the moat, at the moat wall level and above the walkway. This might be linked to potential contamination events such as accidental spillages from the storage pools at the top of the hillock or contaminated ballast used to fill the moat for the construction of the second generation buildings.

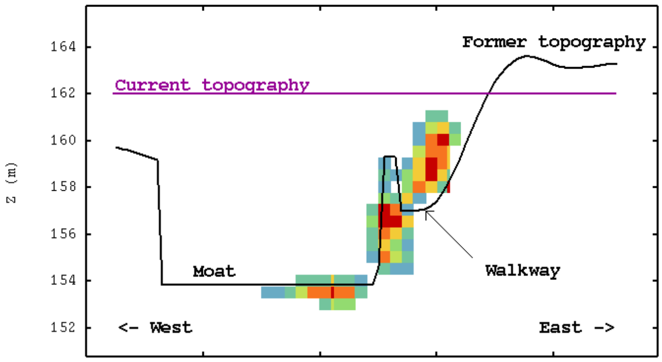

Fig. 7. Transverse section of the contamination, same color scale as Fig. 6. (vertical scale x2).

Encountered Difficulties

During the geostatistical analysis and the interpretation of the results, several issues have been pointed out:

- As the drillhole campaigns were performed by different contractors and with different drilling equipments, the direct comparison of samples is not easy due to their length variability (from 5 cm to 50 cm). A regularization step should be added at the beginning of the geostatistical processing to derive comparable composites of the same size.
- Detection limits are similarly very different according to sample masses and acquisition times that had changed between the first investigation stage and the last one. This also emphasizes the advantage of having homogeneous sampling campaigns without which data pre-processing is compulsory as well as the right choice for threshold that should be above all the detection limits.
- Historical information on the contamination origin is unfortunately unavailable due to the delay between contamination events and recent investigations that represents almost half a century.

CONCLUSIONS

Geostatistical processing proved to be relevant to characterize a deep radiological contamination by the integration of historical data and the advanced spatial evaluation of collected samples from drillholes. Estimation results led to possible scenarios explaining the observed contamination: contaminated ballast and/or contaminated water flooding down to the moat.

Although several drillhole campaigns were not anticipated initially, the iterative sampling strategy turns out to be particularly efficient to adapt the investigations. Indeed, additional drillholes were decided in order to focus on uncertain areas, to delineate the contamination, etc. The contamination interpretation is progressively reinforced and the geostatistical processing is simply updated in this way. On the contrary, the whole collection of required data represents a long period and additional costs for each sampling campaign, which have to be taken into account.

Geostatistical results - selection on probability of exceeding radiological threshold in particular - facilitate the decisions for the remediation process as regards waste removal techniques through cost-benefit analysis (activity versus depth, contaminated volumes and accessibility materials). Remediation works are currently under progress. The contaminated area is divided into six excavation pits. The first two pits dug above the moat succeeded to remove 7 m thick of non-contaminated soils before accessing the thin contaminated layer, which of course has been treated as nuclear waste.

ACKNOWLEDGMENTS

The author would like to acknowledge CEA for providing the dataset used in this work.

All geostatistical calculations and graphics are performed using ISATIS software (Geovariances, 2012).

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