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Analysis of the relationship binding in situ gamma count rates and soil sample activities: Implication on radionuclide inventory and uncertainty estimates due to spatial variability

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ABSTRACT

The paper strives to identify through geostatistical simulations the parameters which build up a correlation between radionuclide activity concentrations measured on core samples and corresponding in situ total gamma count rates measured into boreholes drilled within the contaminated soil. This numerical exercise demonstrates that a linear relationship should exist between logarithmic values of in situ count rates and logarithmic values of activity concentrations when the contamination is strongly structured through space. A sensitivity analysis to some parameters (geostatistical range of the contamination structure, core sampling method, soil water content, multiple gamma-emitter contamination, etc.) is undertaken to identify which situations may impede the use of such a correlation. Then this approach is applied on Chernobyl measurements undertaken in 2015 and compared to the co-kriging method which considers the localization of the measurements and the additional measurements. It appears that co-kriging is a better estimator than linear regression, but the latter remains an acceptable way of estimating activity from gamma emitters and presents better results than lognormal regression. Therefore, total gamma logging measurements performed into boreholes of porous media contaminated by gamma-emitting radionuclides can be used for characterizing contamination and dealing with its spatial variability with the use of co-kriging.

1. Introduction

Recently, for environmental purposes, non-invasive in situ gamma spectrometry techniques have been developed (Benke and Kearfott, 2001; Chesnokov et al., 1999; Clouvas et al., 2007; Kastlander and Bargholtz, 2011; Panza, 2012). The investigation depth is generally restricted to 0.2–0.4 m, depending on the considered soil and radionuclides (Benke and Kearfott, 2001; Chesnokov et al., 1999; Clouvas et al., 2007; Kastlander and Bargholtz, 2011; Panza, 2012). In situ surface gamma spectrometry is therefore well-suited for quantifying radionuclide fallout (Clouvas et al., 2007) and studying the spatial variability of naturally occurring radioactivity (Guagliardi et al., 2013a,b). As it relies on an assumption about radionuclide distribution with depth (Boden et al., 2013; Kastlander and Bargholtz, 2005; Panza, 2012), field gamma spectrometry cannot however take into account the large variability of radionuclide distributions in soils, especially when the source term is buried.

For quantifying gamma-emitting radionuclide inventories in contaminated soils, it is still necessary to get back to invasive methods such as in situ measurements into boreholes (i.e. logging) and measurements on samples retrieved by drilling or coring. Total gamma ray logging has therefore been widely used (IAEA International Atomic Energy Agency, 1979) and remains the most cost-effective in situ technique for detecting uranium (Mwenifumbo and Mwenifumbo, 2013) and transuranic radionuclides (Rohay et al., 2009).

Inferring activity concentrations from in situ gamma logging relies either on a calibration of the gamma probe (Dodd and Eschliman, 1972) or a correlation between sample analyses and in situ measurements (Carlier, 1964).

On the one hand, complex calibration procedures of gamma probes have been developed for characterizing contaminated sites (e.g. McCain et al. (2013)). As they require the use of homogeneous calibration

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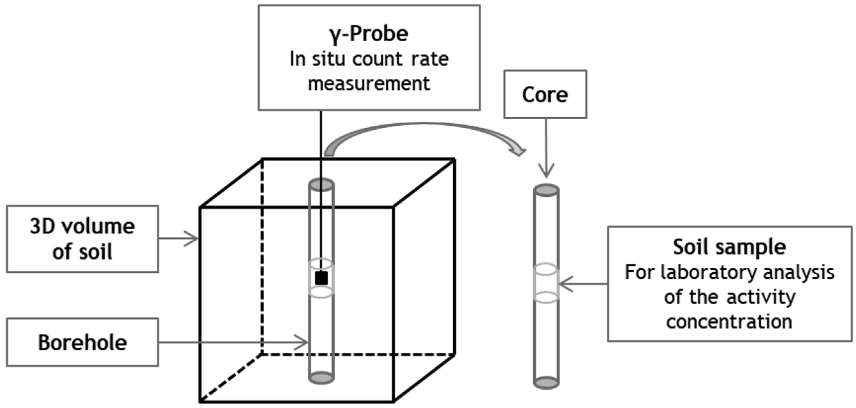

Fig. 1. Principle of the 3D activity model built for studying the relationship between the count rate measured into a borehole with a gamma probe and the activity concentration of a gamma emitter measured by laboratory gamma-spectrometry on a core sample.

models containing mixtures of radioactive minerals (e.g. Stromswold (1994)), the calibrated detector response corresponds to a gamma activity originating from an hypothetic homogeneous uniform distribution of gamma emitters (or gamma-emitting progenies of the targeted radionuclide) within the probe's field of view.

On the other hand, following Matheron (1959) preliminary works in geostatistics, Carlier (1964) studied the relationship between uranium ore grades measured on core samples and total gamma logging measurements performed into the associated boreholes within the rock. He especially focused on estimating the error resulting from the use of a relationship between these variables to infer ore grade estimates from gamma logging measurements. Only a few studies about this correlation have been published so far (Asfahani, 2003; Guillou et al., 2000; Tyler et al., 1996) while it has been extensively used in uranium mining since the 1960s.

Cost-effective characterization techniques providing a good coverage of contaminated zones are required for decommissioning nuclear facilities (OECD & NEA, 2014). Thus the correlation between field gamma spectrometry and soil analysis results has recently been more investigated (e.g. Thompson et al. (2013)). Field data acquired in remediation projects have however still been relegated to supporting analytical data. In some cases field measurements have yet been shown to have some definite advantages over those made in the laboratory due to the uncertainty lying in the sampling procedure (Ramsey et Boon, 2012). Consequently, studying the relationship between in situ count rates and sample activities from a theoretical standpoint is essential to overcome the limited use of in situ measurements in decommissioning and remediation projects.

The paper focuses on revisiting the correlation between radionuclide activity concentrations of soil core samples and the corresponding in situ total gamma ray count rate measured with a gamma probe (lowered into the boreholes at the same depth where soil samples are collected). This correlation is subsequently called the Activity-Count Rate (ACR) correlation.

First, a theoretical exercise is undertaken to identify which parameter physically build up the ACR correlation. It relies on geostatistical simulations of the spatial variability of gamma emitters in a soil volume. A simplified calculation of in situ count rates that would be theoretically measured by a gamma probe lowered in the holes is performed for studying the ACR correlation into the simulated medium. A sensitivity analysis aims to identify the most influent parameters that control the ACR correlation. As demonstrated by previous authors (Guagliardi, I. et al., 2013; Guagliardi et al., 2016a, 2016b), radioactivity measurements in soils are usually influenced by seasonal climatic variations and soil properties. The impact of the moisture content and bulk density on the ACR correlation is therefore explored.

Then, the correlation is studied on measurements deployed on the Experimental Platform in Chernobyl (EPIC) site to test the methodology on a complex case which exhibits a deviation from log-normality. The issues raised by the back-transformation of the correlation established on log-transformed values are explored. In conclusion we suggest some new perspectives on managing inventory uncertainty due to spatial variability.

2. Materials and methods

2.1. Synthetic dataset built from numerical geostatistical simulations

The building of synthetic datasets is first presented together with the geostatistical simulation method.

2.1.1. General principle

A 3D numerical model which represents a field of activity concentrations in soils for a given gamma emitter (e.g. ^{137}Cs) is built. The volume considered is a cube discretized on a regular mesh. An activity concentration (Bq/kg) is assigned to each cell according to a geostatistical model described hereafter. A vertical borehole is then simulated by setting to zero the activity concentration of a column of cells selected at the center of the cube (Fig. 1) for mimicking the removal of material by coring. A count rate is assessed at the center of this borehole as a convolution of the activity concentration in the media surrounding the borehole. The weighted contributions of each surrounding cell decay exponentially as a function of the distance between the probe and the location of the gamma emitters within the simulated medium.

The measured gamma count rate is calculated according to equation (1).

$$F_{insitu}(x) = \int_V A(u) \times \frac{e^{-\mu r(x,u)d}}{4\pi r(x,u)^2} \times \varepsilon \times d \times S \times f(\theta) du \quad (1)$$

Where $F_{in\ situ}(x)$ is a photon flux whose magnitude is proportional to the in situ count rate due to gamma-emitter activities surrounding the measurement point $x = (x_1, x_2, x_3)$ located at the center of the cube. Its dimensions correspond to a number of photons per second perceived at point x through the surface S (m^2) of the probe, equal here to a sphere of one cell size within the borehole; $\int_V$ denotes a volume integral (in each space direction with $du = du_1 du_2 du_3$); $A(u)$ is the activity concentration (Bq/kg) at point $u = (u_1, u_2, u_3)$ within the soil; $r(x, u)$ is the distance (m) between the measurement point x and the point u where the activity concentration $A(u)$ of a gamma emitter is assigned, *i.e.* $r(x, u) = \sqrt{(x_1 - u_1)^2 + (x_2 - u_2)^2 + (x_3 - u_3)^2}$; d is the bulk soil density (kg/m^3) which is considered, at first, constant in space; μ is the gamma attenuation factor (m^2/kg); $f(\theta)$ is the detector angular response of the probe and ε is the gamma-ray emission intensity (%) of the gamma