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Rapid gamma spectrometric analysis of soil samples after radioactive fallout using a "conjugate view" technique

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ABSTRACT

A method for fast determination of the activity in soil samples by measuring a fresh soil core sample from both ends with an HPGe-detector is proposed. The method was tested by simulations, as well as by measurements on spiked soil samples. Both simulations and measurements showed that the geometrical mean of the count rates when the sample is measured twice, with the bottom and the top end, respectively, facing the detector closely resembles the count rate from a measurement on a homogenized sample. Therefore, an efficiency calibration for homogenized samples could be used even if the activity is highly inhomogeneous in the soil core, as is the case with fresh fallout.

It was also shown that by studying the ratio between the two measurements, a rough estimation of the extent of penetration of the radionuclide in the soil can be found. This will enhance the possibility to make rapid estimations of the soil inventory by field gamma spectrometry.

1. Introduction

In the event of an accident involving a release of radioactive elements in the environment, the extent of the release must be estimated in order to take decisions about proper countermeasures. Atmospheric dispersion models are useful tools for estimating air activity and deposition but in order to survey the actual distribution of the fallout, and the activities of the dispersed radionuclides, measurements in the environment is of utmost importance and must be performed, preferably shortly after the release. These measurements can be made by different kinds of mobile or stationary measurements, e.g. airborne gamma spectrometry or field gamma spectrometry, and deposition maps could be produced shortly after the measurement. However, these measurements require equipment that may not be available shortly after an accident and the inventory of radionuclides in the ground must then be determined by soil sampling (Saito et al., 2015). Soil sampling also provides information about the horizontal and vertical distribution of radionuclides (Rosén et al., 1999), data that are necessary when calibrating the mobile systems (e.g. Tyler et al., 1996).

Gamma spectrometry on soil samples is, however, time consuming since several samples must be collected in order to produce a reliable estimation of the deposition. The samples must thereafter be processed before analysis, often including homogenization, drying and sieving. In an early stage after deposition a rapid analysis of the deposition is important in the event of an accident, there is a need for time-efficient methods of sampling and subsequent gamma spectroscopic analysis. These methods should ideally require no processing of the samples prior to analysis while still producing accurate and reliable results. A problem with soil samples from areas with fresh fallout is that the radioactive elements are concentrated in the upper part of the soil core, as well as in the vegetation, while most detector systems are calibrated for homogeneous samples. A direct measurement on a soil sample would then yield a result that deviates from the true activity in the sample. However, the throughput of samples could be greatly increased if it were possible to measure the samples without homogenization.

Our hypothesis is that by making two measurements on a sample, one with the bottom of the sample container facing the detector and another with the top of the container facing the detector, it may be possible to compensate for the activity profile in the sample by applying the conjugate view technique (e.g. Shulkin et al., 1988). If the geometric mean of the two measurements is approximately equal to the result from a measurement on the homogenized sample, the calibration the homogeneous sample

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geometry could be used. The data from the up and down measurements can be verified by homogenizing the sample and performing a third measurement on the same sample. If our assumption is true, the geometrical mean of the counts should coincide with the recorded number of counts for the homogenized sample.

Another hypothesis we have is that the ratio between the count rates obtained from a sample with the bottom of the sample container facing the detector and the same sample with the top of the container facing the detector could give information about the depth distribution of the activity.

The aim of this work was to develop a simple and rapid method to collect soil samples and perform a gamma spectroscopic analysis without preparation of the samples prior to measurement. The measurements performed on a sample could then also be used to determine the depth distribution of the activity.

2. Material and methods

2.1. Determination of the activity in a sample – simulations

The Virtual Gamma Spectrometry Laboratory, VGSL (Plenteda and De Geer, 2002), is a Monte Carlo based simulation platform, which allows the user to model a physical HPGe-detector, lead shielding and source geometry, as well as the composition of the source. VGSL utilizes the Monte Carlo code MCNPX code developed at the Los Alamos National Laboratory, USA.

The geometrical specifications needed for a simulation are crystal radius and length, hole radius and length, and thickness of the dead layers. Dimensions and material of the crystal holder, end cap mantle and end cap window are also given as input data. These specifications were taken from data sheets for to the detector (Ortec Gem 50P4, coaxial, p-type). VGSL also requires an energy calibration, Full Width at Half Maximum (FWHM) calibration and the number of channels in the simulated spectrum. These data were taken from measurements of certified calibration samples. Detector shielding was modelled using the dimensions and material for the actual lead cave used in the measurements.

The simulations reported in this paper do not require an efficiency calibration but since the thicknesses of the dead layers and the distance from the crystal to the end cap are difficult to determine with high accuracy, these parameters were determined by fitting the simulated efficiency curve to an efficiency curve determined from measurements (from about 60 keV to 2 MeV).

The soil samples were an organic model soil with the soil composition of 2.1% H, 1.6% C, 57.7% O, 5.0% Al, 27.1% Si, 1.3% K, 4.1% Ca, 1.1% Fe, simulating a grassland soil. To simulate a soil sample from a fallout area, the modelled sample consisted of a 10 mm thick volume containing the radionuclides (active volume) and a 66 mm thick volume of inactive soil. The diameter of the samples were 70 mm. In order to model the density dependence, one sample, the low-density sample, was composed of an active volume with density 0.50 g cm^{-3} and inactive soil with density 1.50 g cm^{-3}. The high-density sample was modelled with density 1.50 g cm^{-3}, both for the active and the inactive soil volume. The corresponding models for homogeneous samples, acquired by mixing the soil in the two above samples, were thus given the densities 1.36 and 1.50 g cm^{-3}, respectively. The data for the model samples are summarized in Table 1.

Four radionuclides were used in the simulations: ^{131}I (365 keV), ^{137}Cs (662 keV), ^{60}Co (1173 and 1333 keV) and ^{88}Y (898 and 1836 keV). For each sample, a simulation was run with the active volume facing the detector, as well as with the inactive soil facing the detector. The geometrical mean of the count rate at each photon energy from these two simulations was calculated. The geometrical mean was then divided by the count rate from a simulation of a homogeneous sample.

2.2. Determination of the activity in a sample – analysis of soil samples

There were three natural soils collected in the field, at Änggården and Västra Frölunda south of Gothenburg (Lundqvist and Wohlfarth, 2001), 57.67 lat 11.91 long, and two fabricated samples. The soil types and the density from the field soils were: 1) organic soil (including sand and clay), 1.63 g cm^{-3} 2) sandy loamy till, 1.72 g cm^{-3} and 3) forest mor-layer, 1.06 g cm^{-3}. The two fabricated samples were: 4) potting soil, 0.73 g cm^{-3}, which was a peat mixed with sand; pH 6.5 with added NPK, and 5) a pure sandy soil, 1.76 g cm^{-3}.

The soil samples were collected using a newly developed 60 mm diameter and 70 mm length steel core (cylinder) with a handle. The sampler could easily be pressed into the soil by a hammer or simply by the foot. The soil cores taken with this sampler fit smoothly into a 200 mL plastic sample container with screw lid, which was used in this study. The collected soil samples were spiked on the upper surface with ^{99m}Tc, ^{131}I, ^{137}Cs or ^{88}Y, using a small syringe with needle to make the distribution as uniform as possible, and analyzed by HPGe spectrometry (Canberra 229p, efficiency 38%). Only one radionuclide was added to each sample. The added activity was 50 kBq ^{99m}Tc, 15 kBq ^{131}I, 14.4 kBq ^{137}Cs and 29.6 kBq ^{88}Y (Table 1). These activities ensured a statistical uncertainty in the number of counts in the full energy peak below 1%. The activity of ^{137}Cs added to the samples was about three orders of magnitude higher than the activity of ^{137}Cs present in the soil due to fallout from nuclear weapons tests and the Chernobyl accident (Almgren and Isaksson, 2009).

The sample geometry and the detector used in the measurements differ slightly from those used in the simulations. However, both detectors are coaxial, p-type HPGe-detectors of similar size and the source containers have a similar shape and size. The volume of the samples differ by 48% but the ratio height/diameter is similar; 1.1 and 1.2 for the simulation and the measurements, respectively.

Measurements of ^{99m}Tc, ^{131}I and ^{137}Cs were performed on three replicate samples from each of the three types of soil, while measurements of ^{88}Y were performed only on three replicate samples of the organic soil. Each soil core was placed in a plastic bag before being put in the sample container. For each sample, a measurement was made with the spiked surface facing the detector, as well as with the clean soil facing the detector. A new measurement on the same sample was also made after homogenizing the soil within the plastic bag to minimize the risk for contamination of the container. The geometrical mean of the count rate at each photon energy from these two measurements was calculated. The geometrical mean was then divided by the count rate from a measurement of the homogenized sample.

Measurements were also made on fabricated samples consisting of potting soil (density 0.73 g cm^{-3}) and pure sandy soil (density 1.76 g cm^{-3}). Three replicate samples of each type were spiked with each of the radionuclides ^{99m}Tc, ^{131}I, ^{137}Cs or ^{88}Y (Table 1). The activity of each radionuclide was the same as for the sampled soil samples described above.

2.3. Depth distribution – simulations

The depth distribution of fresh fallout with depth can often be described by the exponential relation proposed by Beck (1966):