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Nuclear spectroscopy for *in situ* soil elemental analysis: Monte Carlo simulations

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ABSTRACT

We developed a model to simulate a novel inelastic neutron scattering (INS) system for *in situ* non-destructive analysis of soil using standard Monte Carlo Neutron Photon (MCNP5a) transport code. The volumes from which 90%, 95%, and 99% of the total signal are detected were estimated to be 0.23 m^3, 0.37 m^3, and 0.79 m^3, respectively. Similarly, we assessed the instrument's sampling footprint and depths. In addition we discuss the impact of the carbon's depth distribution on sampled depth.

1. Introduction

Spectroscopy of gamma-rays induced by neutrons is an established technique that can be traced back to when Hevesy and Levi (1936) demonstrated the basic principles of the method using a [Ra/Be] isotopic neutron source to irradiate samples. Since then, analysis of soil samples were carried out using nuclear reactors and n-source systems were developed for soil analysis in the field. For example, *in situ* determination of soil moisture and bulk density was reported by (Gardner et al., 1971; Hignett and Evett, 2002), and elemental analysis in the mining and well logging (oil) industries were reported by (Csikai, 1991; Underwood and Dyos, 1986). At present, soil analysis for carbon, nitrogen, and other nutrients are assessed using chemical analyses; these together with the current state-of-the-art and the newly emerging techniques are reviewed by Chatterjee et al. (2009). Wielopolski et al., (2008, 2010) detail a nuclear technique based on fast neutrons undergoing inelastic neutron scattering (INS) and thermal neutron capture (TNC) reactions. The later uses thermal neutrons resulting from elastic scatterings of fast neutrons with stationary elements present in the soil matrix. The complexities of the INS system and of the soil's matrix, attributable to its multiphase and inhomogeneous composition (Wielopolski et al., 2005), require special considerations when simulating the system response using Monte Carlo Neutron Photon (MCNP) transport code (Breismeister, 1993). We briefly describe the INS system and assess the soil volume it samples using probabilistic simulations. Possible pitfalls when using MCNP and means to circumvent them, when possible, are highlighted.

2. The INS system

Multi-elemental analysis of soil using an INS system is based on spectroscopy of gamma-rays induced by 14 MeV, fast, and thermal neutrons interacting with soil elements via the INS and TNC processes, respectively. An INS system comprises a pulsed neutron generator (NG) operating at frequency of 10 kHz and pulse width (duty cycle) of 25%, an array of NaI gamma-ray detectors with nuclear-spectroscopy electronics, a laptop with software for data-acquisition, and shielding material between the neutron generator and the detectors. The entire system is mounted on a cart 30-cm above the ground and is powered by a 1 kW power generator that when fully operational draws about 1.3 A. The INS system can be operated in either stationary or scanning modes of operation (Wielopolski et al., 2008, 2010). The system was used in various types of fields, ranging from pure organic soils with low bulk density of $\sim$0.5 g/cm^3 to abandoned surface mine fields and forests with soil bulk densities from 1.7 g/cm^3 and up (Wielopolski et al., 2010). The data acquisition system is gated by a NG gate pulse to acquire concurrently two spectra corresponding to INS and TNC signals.

3. MCNP simulations

In the current simulations, we use a time-independent Version 5a of the MCNP code (Pelowitz, 2005); elemental composition that of the soil representing global averages compiled by Frank

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Table 1
The median values of the distributions of the average top thirteen elements from the world soil series; the soil's bulk density is 1.4 g/cm^3.

Element	O	Si	Al	Fe	C	Ca	K	Na	Mg	Ti	N	S	P
Weight (%)	49.0	33.0	7.10	3.80	2.00	1.37	1.36	0.63	0.63	0.46	0.10	0.09	0.08

and Tolgyessy (1993) summarized in Table 1; soil bulk density of 1.4 g/cm^3 and 2% by weight carbon assumed uniformly distributed throughout the soil. It is pointed out that the exact knowledge of the soil composition is not critical and elements below 1% impact the neutron-gamma transport very little, unless they represent strong neutron- or gamma-ray-absorbers. Benchmarking of the simulation is being prepared for publication (O. Doron, private information). The INS response function, R, counts in a peak of interest, is given in Eq. 1;

$$R = k\int_T dt \int_E \sigma(E)dE \iiint_V C_c(x,y,z)\rho_b(x,y,z)\Omega(x,y,z)At(x,y,z)dxdydz \iiint_{V'} Det(E_\gamma,x',y',z')dx'dy'dz' \qquad (1)$$

where T is the time domain, E is the neutron-energy spanning from 14 MeV source energy, down to thermal energies, 0.025 eV, and V is the soil's dimensions, which are semi-infinite in all three directions. V' is the space occupied by the detectors. Thus, for example, the yield of the number of counts in the carbon peak, R, given in Eq. 1 depends on the depth distributions of the various components in the equation. These are:

- $\varphi_n(x,y,z,E_n)$ (n/cm^2) is the calculated neutron-flux depth-distribution; decreases with depth.
- $\sigma(E)$ (cm^2) is the neutron cross-section that only depends on neutron's energy.
- $C_c(x,y,z)$ (gC/cm3), the carbon depth-distribution; it decreases with depth although it may assume different distributions.
- $\rho_b(x,y,z)$ (g/cm^3), is the soil's bulk density; increases with depth.
- $\Omega(x,y,z)$ (fraction), the solid angle subtended by the detectors from the emission point of the gamma-ray; decreases with depth.
- At($E_\gamma x,y,z$) (fraction), the attenuation of gamma-rays on their way to the detector; increases with depth.
- Det(E_γ,x′,y′,z′) (counts in the photopeak) detector parameters and complete energy deposition.

The variables described above require prior knowledge before carrying out MCNP simulations. However, since such data is not always available, the bulk density and elemental concentrations can be replaced with constant values or assumed to be exponentially increasing and decreasing, respectively. For example, in this assessment, the neutron fluxes, gamma-ray attenuation, and the solid angle were calculated as part of the simulations. However, the carbon depth-distribution and bulk density were assumed to be uniformly distributed, in agreement with ones encountered in synthetic soils; 1500 kg samples were placed in a 2.5 × 2.0 cm^2 and depth of 0.50 m pit (Wielopolski et al., 2008). The geometry of the simulation excluding the shielding between the n-source and the detectors is shown in Fig.1.

The yields of gamma-rays intercepting the detectors were calculated from every 2,500,000 one cm^3 soil voxels in the simulated volume. The fluxes were calculated using the F4 tally whereas the detector pulse height distributions were derived with the F8 tally. These were subsequently used to evaluate test volumes as defined below. To accomplish these calculations about 10^9 n-particle histories were used that required about 24 h of calculation time.

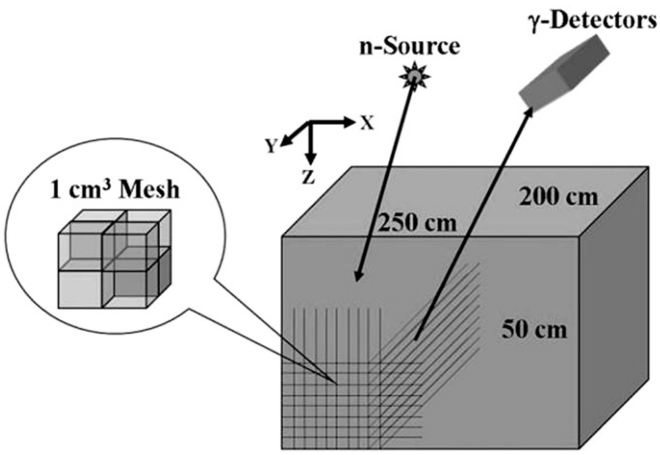

Fig. 1. Depiction of the soil volume's geometry, the n-source, and the gamma-ray detectors used in the MCNP simulations.

Calibration of an INS system presents a particular problem in that the carbon signal measured from an infinite volume is compared to a signal by chemical analysis acquired from a much smaller volume of ~0.0001 m^3. This defies the basic requirements of a calibration procedure in which the reference system and the test system are similar and investigate similar volumes. Clearly, under current practices of correlating INS measurements with those by chemical analysis hardly qualify as a calibration procedure, although they do correlate linearly. Moreover, the real quantity of interest is the elemental surface density, g/cm^2, a quantity that neither of the systems measures directly. In which the surface density is the projection of the volumetric elemental concentration in a defined soil column to the soil surface from a defined depth. These and other MCNP issues are discussed hereafter.

In addition we introduce practical definitions of the sampled volume, test volume, and depth instead of the infinitely large volumes; these dynamic definitions depend on the soil's basic properties, such as carbon, bulk density and depth-distribution. We also discuss the difference between calibrating and correlating the INS in field measurements versus chemical analysis of the soil sample in a laboratory.

4. Results

The infield soil volume sampled by the INS is semi-infinite; therefore, they exceed saturation thickness, which is defined as three mean free paths in which the radiation is attenuated by 99%. Thus, for practical purposes we define an effective test volume from which 90%, 95%, or 99% of the total detected signal is generated. From the simulated soil volume, 2.5 m^3, shown in Fig. 1, we derived the test-volume contours illustrated in Fig. 2. The indentations in the surfaces are due to the shadow shielding between the n-source and the detectors, while the cutoffs on the sides of the 99% contour are due to the narrowness of the sampling box. From the test volumes, we determined the effective sampling depths defined as depth of the apex, the footprints, volumes, and the soil masses; these are summarized in Table 2.

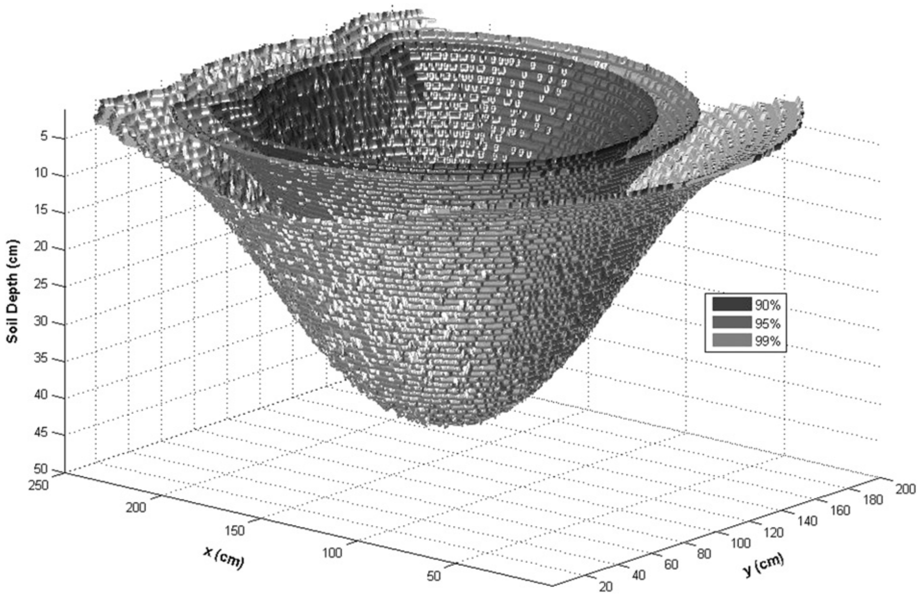

Fig. 2. Contours of the volumes from which 90%, 95%, and 99% of the total detected signal is derived.

Table 2
Estimates of the parameters of sampling depth, footprint, volume, and soil-mass based on the 90%, 95%, and 99% contours.

Total Response(%)	Depth(cm)	Footprint(m^2)	Volume(m^3)	Mass(kg)
90	25	2.4	0.23	326
95	31	3.3	0.37	522
99	44	7.1	0.79	1105

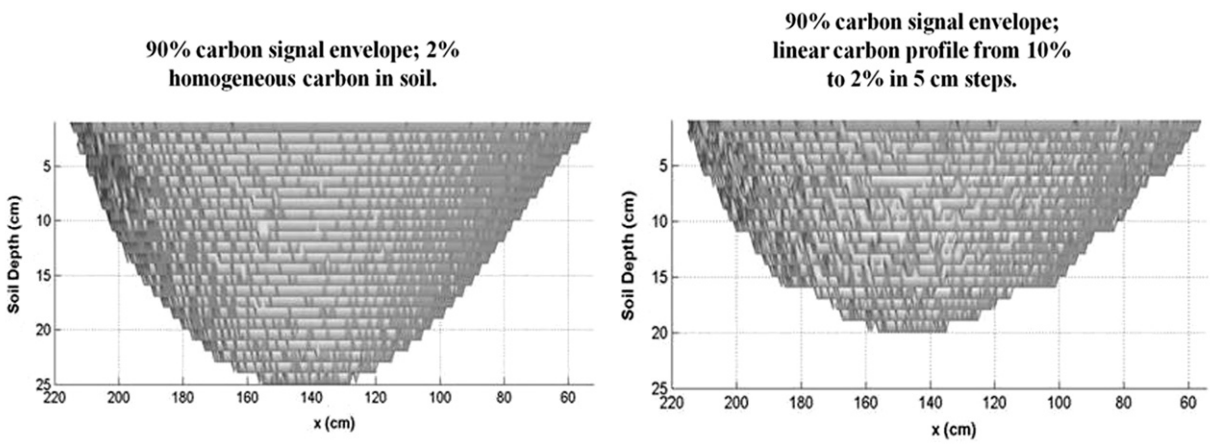

Fig. 3. Lateral views of volumes containing 90% of the total detected signal due to homogeneous carbon distributions in soil (left), and due to a linear one (right).

Thus defined parameters are dynamic and depend on the soils' conditions, the depth distribution of the element of interest, and the soils' bulk density. For example, two lateral views of the 90% contours: a) for homogenous carbon distribution and b) for linear decree with depth of the carbon concentration, are shown in Fig. 3. The decrease in the apex depth from 25 cm to 20 cm with similar footprints clearly results in reduced test volume. We note that despite a reduced test volume, in reality the sampled volume remains the same but the carbon distribution is skewed closer to the surface yielding a 90% value from a smaller volume.

Consequently, the INS system cannot be calibrated in terms of carbon concentration because it varies with depth, and so the effective carbon concentration would depend on size of the test volume. To circumvent this issue, the INS system is calibrated in terms of aerial- or surface- density, g/cm^2, which is projection of the elemental content of a soil column with a known cross-section. This projection must be from a specified depth of the soil column.

Using 1500 kg of synthetic soils, mixture of pure sand with known amounts of charcoal, we plotted the yield of the INS signal against surface carbon density Fig. 4. It shows a good linearity with a regression coefficient r^2 of 0.99. However, due to lack of certified standards it is at best an approximate calibration. The high axis intercept in regression of line "a" in Fig. 4 is due to interference from a cascading 4.45 MeV gamma-ray line from silicone overlapping with that from carbon. Correcting the carbon

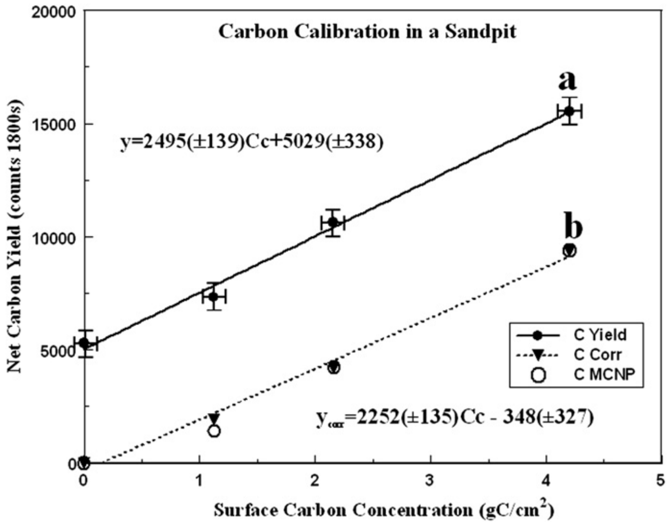

Fig. 4. Regression lines of an INS system using synthetic soils. Regression to a raw data line "a" and corrected for a 4.44 MeV line cascade in silicon "b".

peak for the silicone cascade (Wielopolski et al., 2008) yields a regression line "b" in Fig. 4, with intercept close to the (0,0) point. The circles in Fig. 4 are results of MCNP simulation and normalized at the highest point in regression line "b". The axis intercept at zero due to MCNP calculations suggests that the cross sections used in MCNP calculations lack the silicon's cascading factor of 0.0547 (Herman et al., 2007).

5. Summary

MCNP simulations are critically important in evaluation and characterization of a system enabling analytical simulations of aspects of the system that are difficult or impossible to evaluate experimentally. We used MCNP to assess a newly defined test volume and to derive the footprint with the sampling depth for the INS system. We pointed out that it is next to impossible to calibrate an INS system due to the dearth of large-volume standards. Furthermore, the INS signal correlated linearly with soil surface density, Fig. 4, and it was also validated in field studies indicating proportionality between these two independent variables (Wielopolski et al., 2010). System calibration with synthetic soils revealed the silicone interference of cascading gamma-rays from a 6.23 MeV level to the ground state via a 4.45 MeV level overlapping with the carbon peak. Furthermore, normalizing MCNP results to the experimental data with synthetic soils demonstrated that the standard MCNP cross-sections do not include the silicon-cascading factor.

The pliability of the MCNP to variations in soil parameters we attribute to the averaging processes due to broad beams used and large volume with large angular spread in the radiation transport. For example, we found via simulations and experimentally that variations in the soil's moisture content have little effect on the 14 MeV neutron transport due to reduced hydrogen elastic scattering cross-section for neutrons at energies above 1 MeV and the fact that inelastic neutron absorption with carbon occurs at energies above the threshold energy of about 5 MeV.

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References

Breismeister J.F.. (Ed), 1993. MCNP-A General Purpose Monte Carlo N-Particle Transport Code Version 4A. Los Alamos National Laboratory, Report; NM, LA-12625-M.
Chatterjee, A., Lal, R., Wielopolski, L., Martin, M.Z., Ebinger, M.H., 2009. Evaluation of Different Soil Carbon Determination Methods. Crit. Rev. Plant Sci. 28 (3), 164–178.
Csikai, J., 1991. Nuclear data for geology and mining. In: Qaim, S.M. (Ed.), In: Nuclear Data for Science and Technology. , Spring-Verlag, pp. 644–649.
Frank, V., Tolgyessy, J., 1993. The chemistry of soil. In: Tolgyeddy (Ed.), Chemistry and biology of water, air and soil environmental aspects. Elsevier ISBN 13:978-0-444-98798-3.
Gardner, R.P., Dunn, W.L., McDougall, F.H., Lippold, W.J., 1971. Optimization of density and moisture content measurements by nuclear methods. National Cooperative Highway Research Program, Report, 125.
Herman, M., Capote, R., Carlson, B.V., Obložinský, P., Sin, M., Trkov, A., Wienko, Zerkin, V., 2007. EMPIRE: Nuclear reaction model system for data evaluation. Nucl. Data Sheets 108, 2655–2715.
Hevesy, G.L.H., 1936. Danske Vidensk. Sels. Mat. Fys. Medd. 14, 5.
Hignett, C., Evett, S.R., 2002. Neutron thermalization. In: Methods of Soil Analysis. Physical Methods. Jacob H. Dane and G. Clarke Topp (eds), 501–521 Soil Sci. Soc. Amer.
Pelowitz D.B. (Ed.). 2005. MCNP^{TX} user's manual version 2.5.0.Report LA-CP 05-0389.
Underwood, M.C., Dyos, C.J., 1986. Inelastic neutron scattering reactions in fluid saturated rock as exploited in oil well logging. Appl. Radiat. Isot 37, 475–481.
Wielopolski, L., Song, Z., Orion, I., Hanson, A.L., Hendrey, G., 2005. Basic considerations for Monte Carlo calculations in soil. Appl. Radiat. Isot. 62, 97–107.
Wielopolski, L., Hendrey, G., Johnsen, K., Mitra, S., Prior, S.,.A., Rogers, H.H., Torbert, H.A., 2008. Non-destructive system for analyzing carbon in the soil. Soil Sci. Soc. Am. J. 72 (5), 1269–1277.
Wielopolski, L., Johnsen, K., Zhang, Y., 2010. Comparison of soil analysis methods based on samples withdrawn from different volumes: Correlations versus Calibrations. Soil Sci. Soc. Am. J. 74, 812–819.
Wielopolski, L., Chatterjee, A., Mitra, S., Lal, R., 2011. *In situ* Determination of Soil Carbon Pool by Inelastic Neutron Scattering: Comparison with Dry Combustion. Geoderma 160, 394–399.