Application of Geant4 simulation for analysis of soil carbon inelastic neutron scattering measurements


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HIGHLIGHTS

- The Geant4 Monte-Carlo simulation of inelastic neutron scattering in soil was done.
- The agreement between simulated and measured gamma spectra was demonstrated.
- The carbon INS signal is proportional to carbon content in a ~10 cm soil layer.
- This proportionality is correct for any carbon depth distribution pattern

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ABSTRACT

Inelastic neutron scattering (INS) was applied to determine soil carbon content. Due to non-uniform soil carbon depth distribution, the correlation between INS signals with some soil carbon content parameter is not obvious; however, a proportionality between INS signals and average carbon weight percent in a ~10 cm layer for any carbon depth profile is demonstrated using Monte-Carlo simulation (Geant4). Comparison of INS and dry combustion measurements confirms this conclusion. Thus, INS measurements give the value of this soil carbon parameter.

1. Introduction

The necessity for accurately measuring soil carbon is important since it is a universal indicator of soil quality (Seybold et al., 1997) and can impact many environmental processes, such as soil carbon sequestration, fertility, erosion, and greenhouse gas fluxes (Potter et al., 2001; Torbert et al., 2004; Stolbovoy et al., 2007; Smith et al., 2012). The inelastic neutron scattering (INS) method of soil carbon determination is effective for in situ measurements since it is based on carbon nuclei excited by fast neutrons issuing 4.43 MeV gamma rays promptly after the interaction (Wielopolski, 2011) which can be recorded. The INS system for these measurements consists of a fast neutron source (e.g., a deuterium-tritium neutron generator), gamma detector(s), radiation shielding, data acquisition system, and construction components (Yakubova et al., 2014). The intensity of the carbon gamma signal acquired by the gamma detector depends on the neutron flux intensity and soil properties (density and element content), but the intensity is also strongly dependent on the carbon depth distribution profile. Soil properties determine neutron penetration depth and gamma flux attenuation. The soil carbon depth distribution is usually non-uniform (with carbon content decreasing as depth increases) and can be described, as a first approximation, by exponential law (Wielopolski et al., 2008). The parameters of these distributions vary from site to site (Yakubova et al., 2014). For these reasons, the correlation between the carbon peak intensity in the gamma spectrum and characterization of soil carbon content parameters is not obvious.

A Monte-Carlo (MC) simulation (Hendricks, 1994; Lux and Koblinger, 1991) of these measurements can be conducted to analyze INS measurement results. This mathematical approach is widely used today for solving various problems. In particular, by using MC simulation it is possible to estimate the neutron flux passing through materials and energy lost in the materials (Deiev, 2013), determine the energy distribution of emerging neutrons (Deiev, 2013), calculate the optimal thickness of shielding (Bak et al., 2011; Reda, 2011) and moderator (Feng and Jun-tao, 2014), and reproduce characteristic neutron induced gamma-ray spectra of...
different materials (Guo et al., 2004; Qin et al., 2015; Nasrabadi et al., 2011; Perot et al. 2012). In the present research, MC simulation was used to reproduce the neutron stimulated gamma spectra (both real soil and the sand–carbon mixture) to determine the correlation between the carbon related peak in the gamma spectra and carbon content in the studied object.

The MC simulation model should consist of two major components – the INS system and a soil model. The model INS system should mimic the experimental INS system and may consist of neutron sources (usually an isotropic source with energies used in the experimental setup), detector, and shielding (if needed). The soil model for the MC simulation could generally be considered as a three phase system consisting of solid, liquid and gaseous phases (Wielopolski et al., 2005). Depending on the objectives of the calculations, the soil model need not always be so complicated if all soil parameters critical to the MC simulation are taken into account. In many cases, the soil model can be considered as a compact medium with known elemental composition and density depth profiles (Wielopolski et al., 2005). An MC simulated INS system with a compact medium soil model was used for analyzing the INS system calibration (Doron et al., 2014), optimizing the INS system height above the ground (Doron et al., 2014), estimating the effective sampling volume for detector signal counting (Doron, 2007), analyzing the effect of soil moisture on the gamma signal, and simulating fast neutron transport in soil (Wielopolski et al., 2005). Most of the conducted simulations agreed with experimental result within ~10% accuracy (Doron et al., 2014). To understand the effect of the carbon distribution depth profile on gamma signal values obtained from INS measurements, and to determine the kind of parameter (characterizing soil carbon content) at which the value of the gamma signal is the same for any shape of carbon depth distribution, an MC simulation of these measurements can be conducted. For our purposes, the soil model was constructed as a compact medium with known elemental composition and density depth profiles. The MC simulation represents a step-by-step simulation of randomly issued neutron transportation (taking into account all interactions with soil materials until the neutron loses all its kinetic energy and disappears due to an interaction or reaches the simulation volume boundary) and counts gamma-rays (appearing due to neutron interactions) that pass through the detector volume. A computer screenshot of the MC simulation of neutron propagation through the soil and of gamma rays that appear due to interactions is shown in Fig. 1. Neutrons are emitted (red lines) in random directions from the neutron source, and some penetrate into soil. Due to interactions with soil nuclei, these neutrons lose energy and change direction. Some interactions create gamma rays (white lines) due to various nuclear processes. These gamma rays move through and interact with soil elements. Due to these interactions, gamma quanta lose energy and eventually disappear; however, some leave the soil and pass through the detector (green box) where they are counted. The relationship of gamma counts versus their energy is the simulated gamma spectrum. This spectrum will depend on the soil properties, and each kind of nuclei is responsible for one or several peaks in the spectrum. For instance, carbon nuclei are responsible for the peak with a centroid at 4.43 MeV.

The effect of different carbon depth profiles and soil contents on the gamma spectrum as a whole and on the carbon peak, in particular, can be determined by variation of the carbon depth profile parameters in the model when conducting the simulation. The GEANT4 tool kit (Agostinelli et al., 2003) is suitable for conducting such simulations. Monte-Carlo simulations of 14 MeV neutron propagation in soil samples with different carbon depth profiles and gamma spectra acquisitions were conducted in this work. The purpose of these simulations was to find the soil carbon content parameter for which the value of the INS carbon gamma signal was independent of the carbon depth profile.

2. Materials and methods

2.1. Method of simulation

The Geant4 tool kit (Agostinelli et al., 2003) can be used to conduct MC simulations of the INS soil carbon measurements. The version G4.10.01p.01 (CERN, 2014) was used in this work. A conventional laptop and high performance computing cluster (The Samuel Ginn College of Engineering vSMP HPCC consists of 512 cores @2.80 GHz X5560, 1.536TB shared memory, and 20.48TB raw internal storage) were used for calculation. The simulation model geometry was very similar to our experimental INS system geometry (Yakubova et al., 2014). Specifically, the model consisted of a point isotropic neutron source and gamma detector. The distance between source and detector (35 cm), height of the model system above the ground, and type of detector (NaI(Tl)) were the same as in the real system. Note that, for the accuracy of the simulated spectra to approximately equal the accuracy of the experimental spectra, simulation events ≥ 109 should be performed. On a conventional laptop, the simulation time for this number of events would exceed 200–300 h (depending on the soil model content) with the detector sizes used in our experiments (12.7 cm × 12.7 cm × 15.3 cm). To decrease the simulation time, the model detector was constructed as a cylinder with inner radius \( R_{\text{in}} = 35 \text{ cm} \), and outer radius \( R_{\text{out}} = 50.3 \text{ cm} \), and height=12.7 cm, and the neutron source was placed in the center. This manner of construction decreased the simulation time by a factor of ~20, but due to the equivalence of all detector positions at a distance \( R_{\text{in}} \) from the source, the accumulated spectrum was similar to the spectra simulated with experimental detector sizes. Running a simulation in multitread mode on a laptop with a multi-core processor or on a high performance computing cluster saves significant time. Thus, the spectra simulation time for acceptable analysis accuracy was 4–5 h.

The soil models were constructed based on information about soil element content and density at the measured field sites. This information was taken from chemical analysis or from calibration pit data (sand–carbon mixtures). Further detail about the soil models are discussed later. The neutron cross-section JENDL4.0 database was used in the simulation instead of the default G4NDL4.5 database since the G4NDL4.5 database produced a peak (centroid at 4.43 MeV) that was relatively higher than other simulated spectra peaks compared to the experimental spectra. Analysis showed this may have been due to the \( ^{16}\text{O}(\text{n},\text{n'})\alpha^{12}\text{C} \rightarrow ^{12}\text{C} + 4.43 \text{ MeV} \) reaction cross-section being too large which led to the higher peak value in the spectra. Since the

![Fig. 1. Computer screenshot of an MC simulation of the neutron propagation through the soil. (For interpretation of the references to color in this figure, the reader is referred to the web version of this article.)](image-url)
JENDL4.0 simulated spectra and the experimental spectra were more similar, the JENDL4.0 neutron database was used in this work.

The physics lists QGSP BIC HP and QGSP BERT HP (Reference Physics Lists, 2014) were used. Both physics lists included high precision models for neutron transport below 20 MeV and gave the same simulation results. The change in detector energy resolution was taken into account as \( \sim 1.142 \cdot \sqrt{\langle E \rangle} \) (\( \langle E \rangle \) is the gamma quanta energy, keV) during simulation. This type of energy resolution dependence for gamma detectors is known (Knoll, 2000), and the multiplier 1.142 was found by adjusting the width of the \(^{137}\text{Cs} \) peak in the simulated spectra to match this peak width in the experimental spectra. The detector efficiency dependence with energy was not taken into account since the change would be minor due to the large NaI crystal sizes in the 1–10 MeV energy range (Canberra, 2014).

2.2. Method of measurement

The verification and validation of simulation results was done by comparison with measurement results. The INS system consisted of a pulsed MP320 neutron generator (Thermo Fisher Scientific, Colorado Springs, CO) and three 12.7 cm \( \times \) 12.7 cm \( \times \) 15.3 cm sodium iodide gamma detectors (Scionix USA, Orlando, FL) with corresponding XIA LLC electronics (XIA LLC, Hayward, CA) as previously described in detail (Yakubova et al., 2014, 2015). The Pulsed Fast/Thermal Neutron Analysis (PFTNA) technique (Womble et al., 1995; Barzilov et al., 2012) was applied during measurement. With this technique, two gamma spectra were acquired as a result of the measurement—the so-called inelastic neutron scattering (INS) spectra acquired during the neutron pulse, and the thermo-neutron capture (TNC) spectra acquired between the neutron pulses. INS spectra consisted of gamma rays appearing due to inelastic neutron scattering, thermal neutron capture, and delay activation of nuclei in both the samples and construction materials of the INS system. TNC spectra consisted of gamma rays appearing due to all of the above listed processes, with the exception of the INS process. The “net INS spectra” were restored with channel by channel subtraction of the TNC spectra from the INS spectra (expressed in cps). These represented the spectra of gamma rays appearing due to inelastic neutron scattering in both samples and construction materials of the INS system. The “net INS spectra of a sample” were calculated with channel by channel subtraction of the “net INS spectrum of the INS system” (i.e., INS system background spectrum) from the “net INS spectra”. The INS system background was determined by measuring the INS and TNC spectra in the absence of any large nearby objects (i.e., the INS system was raised to a height greater than 4 m above the ground for these measurements; Yakubova et al., 2014). The “net INS spectra of the sample” received in this manner were used for comparison to the simulated INS spectra and for determining the “carbon peak area” as described below.

2.3. Method for calculating “carbon peak” area

The peak with a centroid around 4.44 MeV in the neutron stimulated gamma spectra for soil or sand-carbon mixtures is the main peak of interest since it correlates with carbon content. This peak is the overlapping of the “carbon peak” (4.43 MeV) and “silicon cascade transition peak” (4.50 MeV) in both the experimental (Wielopolski, 2011) and simulated spectra (Kavetskyi et al., 2015). The part of the peak area with centroid 4.44 MeV associated only with carbon (“carbon peak” denoted as \( \text{Ccorr} \)) was calculated by subtracting the “silicon cascade transition peak” area from the peak area with centroid 4.44 MeV. The “silicon cascade transition peak” area is directly proportional to the “silicon peak” with centroid 1.78 MeV. Coefficient for calculations of “silicon cascade transition peak” from “silicon peak” was calculated as follows. The gamma spectra of sand-carbon mixtures were simulated. Each spectral peak of interest was approximated by one or two Gaussian shape curves using Igor Pro standard software (WaveMetrics, 2013) to determine the area under the curve (Note that, since peak fitting by summing two Gaussians gives approximately the same value as the different components parameter, this sum was used in the analysis, while the area of the components were not used). An example of simulated gamma spectra and fitting peaks with centroid 1.78 MeV and 4.44 MeV by Gaussian shape curves are shown in Fig. 2. We denoted the “silicon peak” area in \( i \)-th spectrum as \( S_{Si} \), peak area with centroid 4.44 MeV in \( i \)-th spectrum as \( S_{C} \), and carbon content in \( i \)-th mixture as \( \text{Cont} \). The assumption was that \( \text{Ccorr} \) can be calculated as \( \text{Ccorr} = \frac{S_{C}-f.S_{Si}}{k} \); \( \text{Ccorr} \) was considered to be directly proportional to carbon content of the mixture (k \( \cdot \) \( \text{Cont} \)) with \( f \) and \( k \) being the coefficients (these designations are shown in Fig. 2 for clarity). Using \( S_{C} \) and \( S_{Si} \) data, the values of \( f \) and \( k \) can be determined by minimizing the expression

\[
\sum_{i} \frac{(S_{C}-f.S_{Si}-k \cdot \text{Cont})^2}{\text{Cont}} \rightarrow \text{min}
\]  

(1)

The \( f \) and \( k \) values were found by equating the derivatives of this sum with respect to \( f \) and \( k \) set to zero. The standard mathematical software, MathCAD (Parametric Technology Corporation, 2013), was used to conduct these calculations.

The obtained \( f \) value from sand-carbon mixtures spectra can be used for “carbon peak” area calculation of any simulated spectra using the following equation:

\[
\text{Ccorr} = S_{C} - f \cdot S_{Si}
\]

(2)

The \( k \) value is a calibration coefficient that can be used in soil carbon content determination for the simulated spectra:

\[
\text{Cont} = \frac{\text{Ccorr}}{k}
\]

(3)

2.4. Comparison of simulated and measured spectra for uniform sand-carbon mixtures

Earlier, a comparison of the dependencies between the net soil carbon gamma signal at neutron irradiation with soil layer thickness from the Monte Carlo simulation and calculations using the gamma response model of Yakubova et al. (2015) were conducted and demonstrated good agreement (Yakubova et al., 2015). Herein, the comparison of results of MC simulated and measured spectra are discussed. The simulations were conducted with model samples of uniform sand-carbon mixtures (0, 1, 2.5, 5, 7.5, 10 w% carbon content) to determine the relationship between \( \text{Ccorr} \) peak area values and carbon content. The simulated gamma spectra are shown in Fig. 3, and fragments of the spectra around 1.78 and 4.44 MeV are shown in Fig. 4. The experimental gamma spectra for the uniform sand-carbon mixtures (0, 2.5, 5, 10 w% carbon content) are also presented in Figs. 3 and 4. Comparisons showed that the simulated and measured spectra were similar. Some differences between the measured and simulated spectra can be explained by the simplicity of the model and not accounting for the detector efficiency with energy. While the measured and simulated spectra have insignificant discrepancies, the main features (i.e., position and relative intensity of pair production; and silicon, oxygen, and carbon peaks) are approximately the same.

The dependencies between the peak areas with centroid around 4.44 MeV, “silicon peak” with centroid 1.78 MeV, and \( \text{Ccorr} \) with carbon content from simulated and measured spectra are presented in Fig. 5. As can be seen, the dependencies in both cases are similar to each other. In addition, the coefficient \( f \) values from
data processing of both the experimental and simulated spectra are very close (0.054 and 0.058, respectively). Values of this parameter, which is the coefficient of the cascade transition for $^{28}\text{Si}$ nuclei, are close to earlier published values (Herman et al., 2007; Kavetskiy et al., 2015). This fact can be considered as verification of our MC simulations. The agreement between our MC simulation with results of another calculation method verifies the reliability of our conclusion.

2.5. Soil with a non-uniform carbon depth profile

The samples used in the simulation of spectra with non-uniform carbon depth profile were 200 cm $\times$ 200 cm $\times$ 60 cm. Figs. 6–8 show carbon depth profiles, soil density examples, and main element depth profiles used in the simulations. These carbon depth profiles were from the applied field (AF) at the Piedmont Research Unit, Camp Hill, AL, USA where the measurements were conducted. Fig. 7 shows an example soil density depth profile, and Fig. 8 shows an example depth profile for the main elements present in the AF soil. All represented data were obtained from
chemical analysis of cores taken at the AF sites and were used for soil model construction in the simulation. Six artificial carbon depth profiles with extremal shapes (Art1-Art6 in Fig. 6) were used in the simulations as well.

3. Results and discussion

The purpose of this research was to determine an unique soil carbon content parameter (independent of the carbon depth profile shape) that correlates with INS results. In general, the average carbon content or integral by some depth can be used to characterize carbon content dependence with depth. The tested candidates were: average parameter – average weight percent carbon in some soil layer \([\text{AvgW}\% (h)]\), where \(h\) is the layer thickness\] and integral parameter – grams carbon

Fig. 5. The dependencies of the peak areas with centroid around 4.44 MeV and 1.78 MeV, and \(C_{corr}\) with carbon content for simulated and measured spectra from sand-carbon mixtures.

Fig. 6. Soil carbon depth profiles for sites (see text for details) used in simulations.

Fig. 7. An example soil density depth profile used in simulations.
per square centimeter of soil surface in a layer of some thickness [SD(h), surface density]. These parameters can be calculated as:

\[
\text{AvgW\%}(h) = \frac{1}{h} \int_0^h W\%(b) \, db
\]

\[
\text{SD}(h) = \int_0^h W\%(b) \, db
\]

where \(W\%(b)\) is carbon weight percent at depth \(b\) and \(d(b)\) is the soil density at depth \(b\).

Initially, the calibration dependencies for uniform sand-carbon mixtures were constructed. For this, the coefficients \(f\) and \(k\) were determined as described above for both cases where \(\text{Cont}\), corresponded to \(\text{AvgW\%}(h)\) or \(\text{SD}(h)\). For uniform mixture, \(\text{AvgW\%}(h)\) does not depend on \(h\) and equals \(W\%). Thus, there is only one set of coefficients \(f\) and \(k\). SD depends on \(h\); therefore, each \(h\) has its own set of coefficients. In the set of coefficients for SD, the coefficient \(f\) is the same for each \(h\) and approximately the same for \(f\) for weight percent. Using the determined sets of coefficients, the dependences of the “carbon peak” area (\(\text{Ccorr}\)) in the simulated spectra versus carbon weight percent (\(W\%\)) and versus carbon surface density at different thicknesses [\(\text{SD}(h)\)] in sand-carbon mixtures samples were plotted (Figs. 5 and 9). As shown in these figures, the dependences of \(\text{Ccorr}\) with \(W\%\) and with \(\text{SD}(h)\) are directly proportional within simulation accuracy limits for all cases. After determining the peak areas with centroids at 1.78 MeV and 4.44 MeV, the “carbon peak” area (\(\text{Ccorr}\)) using coefficient \(f\) can be calculated for any soil gamma spectrum. Finally, the \(\text{AvgW\%}_{\text{calc}}\) and \(\text{SD}_{\text{calc}}(h)\) values for each spectrum were determined using coefficient \(k\) from calibration dependences. For sites with uniformly distributed carbon, these values will be equal carbon for weight percent \(W\%\) and surface density \(\text{SD}(h)\). For non-uniform soil carbon distribution the question remains: which parameter would correspond to the values calculated as described above? To solve this problem and to determine the carbon content characterizing parameter that correlates with INS results for any carbon depth distribution pattern, simulations of INS spectra for the AF and Art sites were conducted (using data from Figs. 6–8) along with recording of the simulated gamma spectra (Fig. 3). Peak areas with centroids at 1.78 MeV and 4.44 MeV, and \(\text{Ccorr}\) were calculated. Next, the values of \(W\%_{\text{INS}}\) and \(\text{SD}_{\text{INS}}(h)\) were calculated using sets of coefficients as determined above. In addition, \(W\%_{\text{calc}}(h)\) and \(\text{SD}_{\text{calc}}(h)\) were calculated (Eqs. (4) and (5)) from known carbon depth distributions and soil density depth profiles for these sites (Figs. 6–8).

The relative difference between \(W\%_{\text{INS}}\) from the simulated spectra and \(\text{AvgW\%}_{\text{calc}}(h)\) values from soil model data was used to compare these values. The relative difference \(\frac{|\text{AvgW\%}_{\text{calc}}(h) - W\%_{\text{INS}}(h)|}{W\%_{\text{INS}}(h)}\) for each site and with \(h\) were calculated and plotted in Fig. 10. This relative difference was found to equal zero at some layer thickness \(h\) for each site. Therefore, the results received from INS simulation (value of \(W\%_{\text{INS}}\)) can be attributed to average carbon weight percent in the soil layer with thickness \(h\). As shown in Fig. 10, this depth varied around 10 cm in the range of \(\pm 2\) cm for all sites. This variation is at the limit of peak area determination accuracy for INS simulations. Since different carbon depth profiles (constant to sharp drops) were used in the simulations, the parameter (average carbon weight percent in soil layer with thickness 10 cm) can be assigned the value determined from INS gamma spectra. In contrast, the values \([|\text{SD}_{\text{INS}}(h) - \text{SD}_{\text{calc}}(h)|]/|\text{SD}_{\text{INS}}(h)|)\) were not found to equal zero for any \(h\) for all sites.

The sum of relative differences between values (calculated from INS simulated spectra and soil model data for different sites) can be used to compare different types of soil carbon content parameters (i.e., average weight percent and surface density). These sums for weight percent and surface densities can be calculated by Eqs. (6) and (7), respectively:

\[
\xi_{\text{AvgW\%}}(h) = \sum_n \frac{|\text{AvgW\%}_{\text{calc}}(h) - W\%_{\text{INS}}(h)|}{W\%_{\text{INS}}(h)}
\]

\[
\xi_{\text{SD}}(h) = \sum_n \frac{|\text{SD}_{\text{calc}}(h) - SD_{\text{INS}}(h)|}{SD_{\text{INS}}(h)}
\]

where \(n\) is the site number (the simulation was done for 15 sites).

Equality \(\xi_{\text{AvgW\%}}(h) = \xi_{\text{SD}}(h)\) to value zero means that, at this \(h\), the soil carbon characteristics received from the spectra and depth distribution are the same. The \(\xi_{\text{AvgW\%}}(h)\) and \(\xi_{\text{SD}}(h)\) dependencies for AF and Art are shown in Fig. 11. As can be seen, the carbon weight percent calculated from the spectra coincides with the average weight percent at a thickness of \(\approx 10\) cm. As also shown in Fig. 11, the values of surface density from the spectra and from depth profiles differ from each other at any thickness. Based on these results, we can conclude that the carbon soil content parameter (determined from the gamma spectra using calibration data from a uniform carbon-sand mixture) is the average carbon weight percent for a 10 cm soil layer. This parameter can be used to characterize soil carbon content for any carbon depth distribution shape.

Using the above described approach, analyses of experimental INS gamma spectra were conducted at field sites (Yakubova et al., 2015). Comparisons of carbon average weight percent (upper 8–10 cm layer) from measured INS gamma spectra demonstrated good agreement with values from standard chemical analyses. Thus, we conclude that the carbon average weight percent in \(\approx 10\) cm upper soil layer can be accurately measured by this INS method.
4. Conclusion

INS gamma spectra of soil models (consisting of uniform sand–carbon mixtures) were simulated using the Geant4 toolkit. Using simulation results, calibration lines were generated for ‘carbon corrected peak area’ (with centroid at 4.43 MeV) versus carbon weight percent and versus carbon surface density with different layer thicknesses in mixtures. The INS gamma spectra of soil models with different carbon depth profiles were also simulated. Using carbon corrected peak areas and calibration lines, the values of carbon content characterization parameters (carbon weight percent and carbon surface densities) were calculated. The average carbon weight percent and surface density for layers with different thicknesses were determined from soil model data (carbon depth profiles and soil density depth profiles). Based on the comparison of carbon soil characterization parameters defined from INS gamma spectra and soil model data, it was shown that the parameter value defined by INS gamma spectra can be attributed to the average carbon weight percent in the upper soil layer (~10 cm thickness) for any carbon depth distribution profile. This conclusion was confirmed by field studies that showed good agreement between our technique and the standard dry combustion method. Thus, INS measured gamma spectra using calibration data from uniform sand–carbon mixtures can accurately measure the average carbon weight percent in upper ~10 cm soil layer.

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