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Basic considerations for Monte Carlo calculations in soil

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Abstract

Monte Carlo codes are extensively used for probabilistic simulations of various physical systems. These codes are widely used in calculations of neutron and gamma ray transport in soil for radiation shielding, soil activation by neutrons, well logging industry, and in simulations of complex nuclear gauges for in soil measurements. However, these calculations are complicated by the diversity of soils in which the proportions of solid, liquid and gas vary considerably together with extensive variations in soil elemental composition, morphology, and density. Nevertheless use of these codes requires knowledge of the elemental composition and density of the soil and its physical characteristics as input information for performing these calculations. It is shown that not always all of the soil parameters are critical but depend on the objectives of the calculations. An approach for identifying soil elemental composition and some simplifying assumptions for implementing the transport codes are presented.

Keywords: Monte Carlo; Soil elements; Neutrons; Gammas; Transport

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1. Introduction

The extensive increase in the computational power of the desktop computers facilitated a widespread use of Monte Carlo codes for gamma-neutron transport calculations. These are probabilistic codes that simulate step-by-step the processes that guide radiation transport in matter. These codes enable complex system simulations which other-way would be very time consuming, costly, and occasionally impossible to perform. These simulations can be performed with an arbitrary degree of details provided that all the basic data and complete system description are available. One of these codes is the Monte Carlo Neutron Photon (MCNP) transport code, developed in Los Alamos National Laboratory (Breismeister, 1993), that has been widely used for radiation transport in soil. For example, MCNP has been used for evaluating soil activation, radiation shielding in soil, well logging industry, and for design of complex gauges based on nuclear techniques. One such technique, which gained widespread use, is in situ, non-destructive, multi-elemental analysis of bulk geological samples using neutrons (Csikai, 1991; Clayton and Coleman, 1985). MCNP codes require an input file that contains complete information about the radiation source, the geometry of the simulated system, and both the density and the elemental composition of all materials through which the radiation passes. In addition the output tallies to be calculated need to be specified.

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Due to high variability and inhomogeneous nature of soils, special care must be taken when implementing MCNP simulations. The main contributing factors to complexity in using MCNP for in-soil calculations are; (1) soil is a three-phase system in which the solid phase contains organic and inorganic components, (2) the liquid phase carries many different solutes depending on soil mineralogy, (3) the gaseous phase, although generally ignored, may contain gases that strongly absorb neutrons, (4) on a micro-scale soil is a highly inhomogeneous matrix, although on macro scale some homogeneity and uniformity can be assumed, and (5) soil density primarily depends on porosity and water content. These factors are further complicated by the nature of the geological formation being studied, weathering conditions, and depth. For example, it is well established that the composition of a formation at depth differs significantly from that on the surface. These factors make it nearly impossible to provide exact descriptions of the system. Introducing simplifying assumptions that depend on the degree of accuracy required by the final objectives of the calculation to overcome these complexities. However, the validity of these assumptions must be tested and validated either by conducting simple experiments or by performing analyses of the sensitivity of the calculations to perturbations in the assumptions.

Two essential parameters required for MCNP calculations are soil density and elemental composition. These two primary properties, and an assumption about soil uniformity are pivotal when designing gadgets for soil moisture and porosity measurements (Gardner et al., 1971). These properties are also required when gamma spectroscopy of neutron induced gamma radiation, due to delayed, prompt, or inelastic neutron interactions, are used to solve geophysical problems for the well logging industry (Schweitzer et al., 1993; Grau et al., 1993), or when performing non-invasive in situ quantitative analysis of the elements present in soil (Clayton and Coleman, 1985).

We describe the use of Monte Carlo calculations applied to modeling inelastic neutron scattering (INS), a process we are developing as a technique for quantification of carbon in soil. INS analysis of soil carbon can be carried out rapidly (30–60 min) and does not disturb the soil column. This permits re-sampling at the identical spot over time. Each observation provides a value for approximately 75–100 lb of soil. This is large enough to average much of the small-scale heterogeneity in soils (Wielopolski et al., 2000). At present soil carbon is measured by taking soil samples to the laboratory. This conventional process of extracting soil cores disrupts the soil column and the area around the sampling point. Alternative methods for soil sampling, laser induced breakdown spectroscopy (LIBS), (Cramers et al., 2001; Kincade, 2003) and infrared spectroscopy (McCarty et al., 2002). However, these in situ approaches are invasive. In the first case, small volumes of about 50 μl are vaporized and emission spectroscopy is performed in the second case it is basically surface analysis; these methods are therefore not comparable to the INS method. With re-sampling of the identical spot, and averaging over a relatively large volume, small changes in soil carbon content may be detected that, using conventional methods, would be obscured because of soil heterogeneity. For example, for till no till agriculture (Lal et al., 1999) scanning large areas for carbon content will be very useful to assess the changes in carbon content.

2. Soil components

As pointed out the essential soil parameters, in addition to the geometry, required for Monte Carlo simulations are the elemental composition and the bulk density. These are required to calculate the macroscopic transport cross section for neutrons and gamma radiation, i.e., the probability of interaction with the individual elements present in the matrix, and the density is required for finding the number of atoms in a volume of interest and calculating the range of travel for the radiation.

However, the variety of soil types and the multiplicity of their conditions make it but impossible to obtain a simple parameterization descriptive of all soils. Instead, a semi-systematic approach is used to describe the density and composition of the soil surface and/or near surface, down to about 100 cm. In this region the soil is a loose mixture resulting from physical and chemical weathering and biological processes. It is a heterogeneous system consisting of solid minerals, organic matter, and liquid and gaseous components. The proportion of each component varies largely from site-to-site and depends on the climate and stage of soil

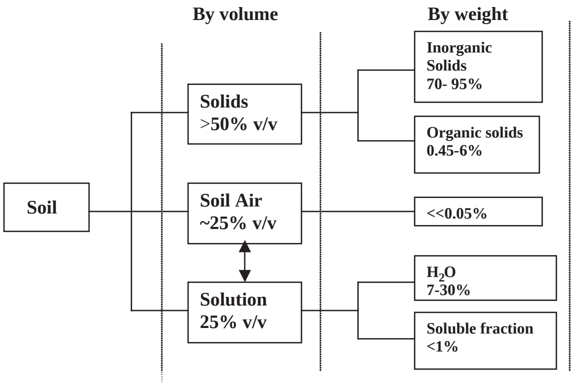

Fig. 1. Soil components with fractional distribution by volume and weight.

development. For example, water content strongly depends on the soil porosity and climate, which in turn affect soil weathering and other processes in the soil. General proportions of the soil main components are shown in Fig. 1, those include the three basic phases namely; solid, liquid and gaseous, in which the solid phase is further subdivided into inorganic and organic components and the liquid contains water and solutes. The relative abundance by volume and by weight of the different compartments is also indicated in Fig. 1.

2.1. Solid mineral component

Solid minerals in soil may make up to 95% of the soil material and determine many of the soil properties. The simplest soil inorganic classification is by particle size distribution. The simplest method of characterizing soil solid is granulometric analysis in which soil solid parts are classified according to the particle size ranges. These size ranges are; gravel, >2.0 mm, coarse sand, 2.0–0.2 mm, fine sand 0.2–0.02 mm, silt 0.02–0.002 mm, and clay, <0.002 mm (US Department. of Agriculture, 1951). Particle-size distribution of soil varies greatly from place to place and also with depth. For example, Table 1 shows the size distribution of three common types of soils in the United States (Barshad, 1964). Although this classification does not reveal any information about the chemical composition of the soil it is often an indication of the mineral components in a well-developed soil. For example, particles with size larger than 0.01 mm consist mainly of quartz, while particles smaller than 0.01 mm are by-and-large clay minerals.

Minerals in soil are generally divided as primary and secondary minerals and they can make up to 99% by weight of the solid component of the soil. Mineral abundance would depend on the types of original rocks on the earth surface and the degree of weathering. Minerals initially derived from igneous and metamorphic rocks constitute the primary minerals that decompose during soil formation process into secondary minerals following a stability order as indicated in Fig. 2. The mineralogy and crystallographic structure of these primary and secondary minerals is well defined and characterized. For example, well developed soils may contain numerous kinds of minerals, however, they will be dominated by quarts which might be present in the 50–90% by weight of the sand and coarser silts. In addition, sodium feldspar, e.g., albite, potassium feldspars and micas, although very resistant to weathering, may occur in much lesser quantities in soils in temperate climatic zones. Since the chemical composition of each mineral is known based on fixed stochiometry, estimates of the elemental composition of a given soil could be derived. Partial breakdown of the soil components is qualitatively depicted in Fig. 3 with additional breakdown of the inorganic solid matter shown in Fig. 4. Elemental composition of some of the soil minerals is given in Table 2.

2.2. Organic component

Organic components may make up to 7% of soil mass. The main source of organic matter in soil is from biota and metabolic activities in the rhizosphere, including root growth and natural decay of the root

Table 1
Particle-size distribution of clay and non-clay soils with uniform depth distribution

Depth (in)	% of whole soil							
	Fine gravel	Sand				Silt	Clay $<5\,\mu$	Total
		Coarse	Medium	Fine	Very Fine			
Greenfield sand								
0–15	8	19	11	24	21	10	6	99
15–40	9	13	12	23	24	12	6	99
40–72	8	20	11	23	20	12	6	100
Hannaford Sandy Loam								
0–16	8	16	8	18	20	18	11	99
10–72	8	16	7	19	19	19	11	99
Montezuma Clay Loam								
0–14	—	0.0	1.3	1.2	8.7	32.7	28.1	72
14–32	—	0.0	0.1	0.1	8.5	35.2	29.1	73
32–40	—	0.7	0.1	1.0	8.1	34.8	29.0	73.7

systems, soil heterotrophs and from plant litter on the surface. Similarly to inorganic matter, soil organic matter also shows a great deal of variability in its chemical composition. For example, a mature dry plant tissue consists of: (1) Carbohydrates with sugars and starches 1–5%, hemicelluloses 10–28%, and cellulose 20–50%, (2) Fats, waxes, tannins, etc., 1–8%, (3) Lignin 10–30%, and (4) Proteins (simple water soluble and crude proteins) 1–15%. Organic solids often coat or aggregate with inorganic particles into soil structure and they constitute most of the carbon in soil. Nitrogen is also contained in the organic matter and may range from 0.02% to 0.25% by weight of the soil make-up; it is about 5% of the total organic matter. Table 3 provides some general distribution of various chemical components of the living and non-living organic matter in soil; the elemental composition of the organic matter is given in Table 4. The composition of the green tissue of high plants consists of 75% or more of water, 11% of carbon, 10% of organically bonded oxygen, 2% of hydrogen and about 2% of ash (Sposito, 1989). More detailed information about the elemental composition of soil organic matter is presented in Table 3. In soils carbon is about one seventh of the organic matter content while nitrogen is about one twentieth, the C/N ratio varies

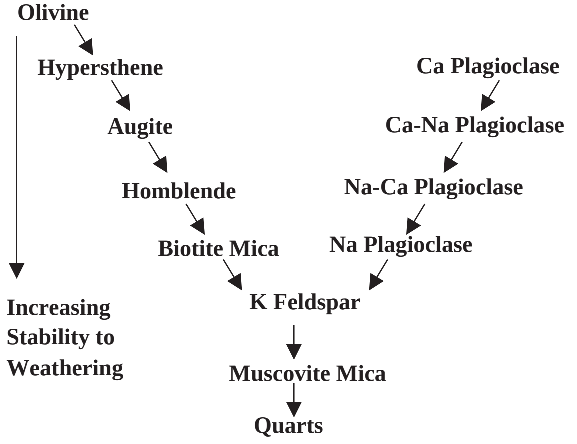

Fig. 2. Stability order of some primary igneous and metamorphic minerals under the surface weathering conditions.

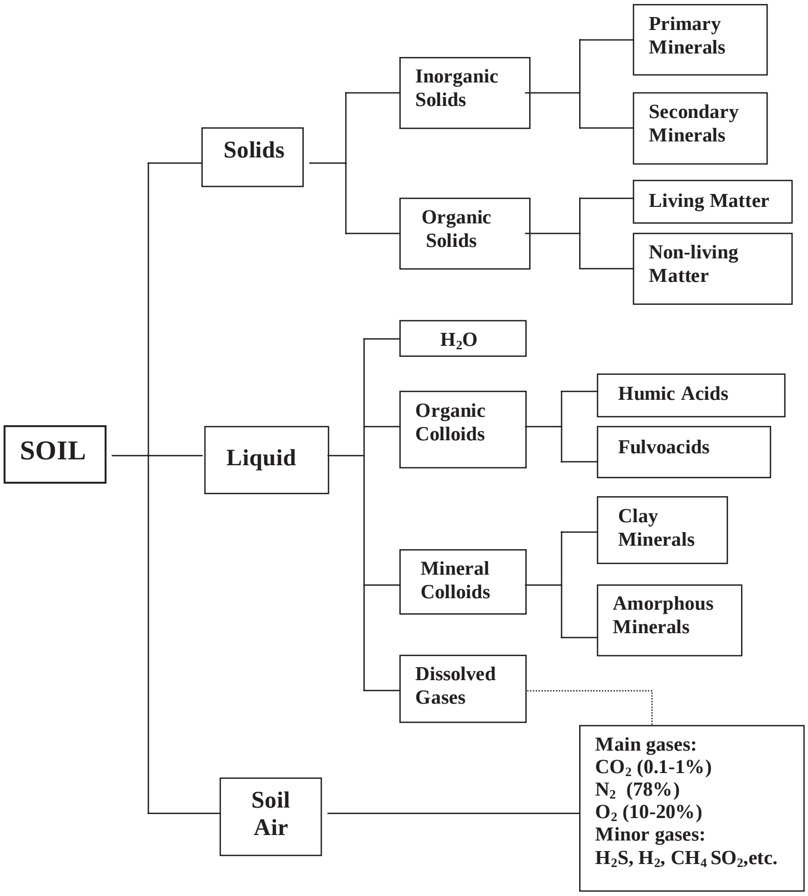

Fig. 3. Partial breakdown of the soil main components.

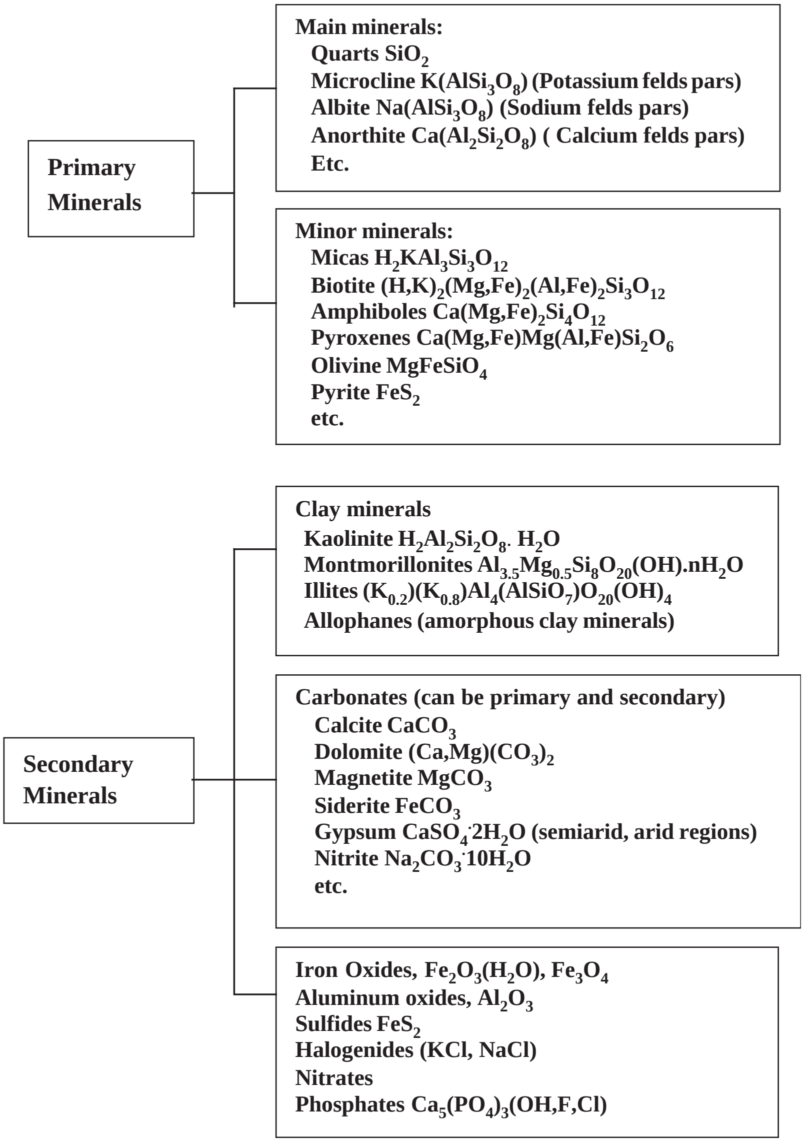

Fig. 4. Partial breakdown of the primary and secondary minerals.

typically from 8:1 to 15:1, (Buckman and Brady, 1969). Therefore, for a given soil, the knowledge of organic carbon content is equal to that of organic matter content. Qualitative breakdown of the organic matter is shown in Figs. 3 and 5.

2.3. Liquid

Soil water, located in the soil pore space, together with the dissolved salts, dissolved organic matter, gases and dispersed substances from various source is defined as the soil solution. This is the most dynamic constituent of the soil. Soil solution is not only important in supplying the nutrients to growing plants but also serves as the carrier for the elements that exchange among the different soil components in the various phases. Knowledge of the water content in soil, and more specifically the amount of hydrogen in soil is important because; (1) hydrogen will absorb very effectively thermal neutrons thus reducing the flux and (2) hydrogen is the best moderator for fast neutrons thus slowing them down and reducing the depth penetration of the neutrons.

2.4. Air

The gaseous phase in soil occupies the pore space and is referred to as the soil air. The main constituents of the soil air are; CO_2, H_2O (vapor), O_2, and N_2, while other

Table 2
Major elemental content of common soil minerals

	Minerals	Si	Al	O	Ca	Mg	K	Na	C	S
Primary Minerals	Quartz	46.7		53.3						
	Microcline	30.3	9.7	46.0			14.0			
	Albite	32.1	10.3	48.8				8.8		
	Anorthite	20.2	19.4	46.0	14.4					
	Pyrite									53.3
Secondary Minerals	Kaolinite	22.7	25.6	51.7						
	Montmorillonites	33.6	14.2	50.4		1.8				
	Illites	4.9	19.9	69.7			5.5			
	Carbonate			48.0	40.0				12.0	
	Iron oxides			28.0						
	Dolomite			52.3	21.7	13.0			13.0	
	Phosphates	19.2			41.2					

gases such as H_2S, H_2, CH_4, SO_2 and many other gases can be encountered, and their presence will depend on the soil conditions. Although by volume the soil air might occupy a large fraction, its contribution to chemical composition by weight is negligible as indicated in Fig. 1. Although very unlikely, nevertheless, attention must be paid to whether the soil air contains strong neutron absorbers such as: B, Li, Cd and Gd, these gases could affect the MCNP calculation significantly and therefore have to be carefully evaluated even on trace level, for instance few tens of ppm.

3. Soil density

Soil density, also referred to, as bulk density, D_b, is a critical soil parameter required for the MCNP calculations. Soil bulk density is defined as the total soil weight per unit total volume. Inherent in this definition is the assumption that the soil's three-phase system has been homogenized, which is a reasonable assumption as long as the soil particle sizes are smaller than the neutron or gamma rays mean free path. The stated importance of the bulk density is due to the fact that the neutron macroscopic cross sections and gamma mass attenuation coefficients depend on it. Thus the bulk density will affect the reaction rates and penetration depth of neutrons and gamma radiation. When combined with the elemental composition of the soil it will also affects the slowing down process (thermalization) of the fast neutrons.

Soil density depends on soil morphology, i.e., particle shape and size distribution, which is characterized through the porosity parameter, S. Porosity is defined as the ratio of the nonsolid, pore space divided by the total space. The pore space may be partially, P_v, or completely, $P_v = 1$, filled with water thus affecting the soil density, where P_v is defined as the water volume in the pore space divided by the total pore volume. If the density of the solid matter is assumed to be D_s and that of the liquid phase D_w, then the soil bulk density can be expressed as:

$$D_b = D_s(1 - S) + D_w S P_v. \qquad (1)$$

For simplicity, assuming arbitrarily a density for the solid and liquid components to be 2 and 1 g/cm^3, respectively, and a range of values between 0 and 1 for both P_v and S, Eq. (1) is depicted in Fig. 6. However, more realistic values for these parameters are for D_s, 2.4–2.7 g/cm^3, D_w slightly above 1 g/cm^3 because of the dissolved minerals, porosity 25–50%, and D_b 1.2–1.6 g/cm^3 (Frank and Tolgyessy, 1993; Tolgyessy, 1993). The significance of Eq. 1 is that it does not exhibit sharp gradients in the bulk density in soil. Those can be encountered at the interfaces between soil and solid rocks. However, as long as the volume with substantial changes in the soil density remains small relative to the beam size there will be an averaging effect that will smooth out these sharp discontinuities (Schacklette and Boerngen, 1984; Bowen, 1979; Miller and Turk, 1951; The Nanking Institute of Soil Science, 1982).

4. Soil elemental composition

In the scheme that we propose, once the components of the soil have been identified it is possible, based on the knowledge of the individual stoichiometries, to derive the elemental composition of the soil, or at least to identify the mean concentrations of the major elements O, Si, Fe, Al, K, and Ca dominate soil elemental composition that is mainly determined by the minerals. Occasionally the exact knowledge of an abundance of an element needs to be replaced with empirical ratios such as C/H = 7.6, C/N = 12, C/S = 70, C/P = 50, C/O = 1.87. If no information about the soil is available then few

Table 3
Chemical composition of soil organic matter (dry weight percent)

		Protein	Lipids (e.g. fats, waxes, tannins)	Carbohydrates			Lignin	Ash
				Cellulose	Hemi-celluloses	Others Sugars and Starches		
Living matter (5–25%)	High plants	1.2–1.6	0.7–3.2	40.0–47.5	23–30		22–30	0.5–1
	Bacteria	50–80	10–30	5–15				
	Grass	5–15	3–10	30–50			10–30	
Non-living (75–95%)	Humin	Humic substances are the products of polymerization of all these components, and the ratio of HA/FA varies from 0.14 to 1.96 in various soils.						
	Humic acids							
	Fulvic acids							
	Dry plants	1–15	1–8	20–50	10–28	1–5	10–30	

samples of the soil can be subjected for elemental analysis. An example of results from soil analysis is shown in Table 5, Soil A for which the density used was 1.59 g/cm^{-3}. In this case there was a need to determine soil activation due to high-energy neutrons, thus trace level elements were also included. Soil B in Table 5 represents a carbon rich loam soil used by the PEGS5 Monte Carlo code for transport calculations in soil (Nelson et al.). Median values of world soils (oven-dried) are given in Table 5 (Sumner, 2000). Since the true distribution of the elements in the soil is seldom known a uniform distribution is frequently assumed although it is well known that there are sharp gradients in C, N and D_b from the soil surface through the first half-meter of the soil profile.

5. Sensitivity analysis

Sensitivity analysis enables testing the validity of the assumptions and or approximations made during the simulations. Typical assumptions may refer to elemental composition, homogeneity, or soil density. Introducing a change, small or large depending on the effect, in the parameter of interest in the input file and observing the variations in the results provides information on the sensitivity of the simulation to this particular parameter. Large variations in the results are indicative of the criticality of the given parameter to the overall simulation. There are no clear rules what constitute large or small variation and it is up to the investigator to make a judgment call.

For example, when calculating soil activation detailed elemental composition of the soil is important. The different activation cross-sections and half-lives for each element affect the total induced activity and the subsequent decay rate. This is demonstrated how the radioactivity is at the end of the irradiation period, short term, and after cooling time, long term, depends on the elemental composition of the soil. However, it is also shown that the neutron penetration in the soil is less dependent on the elemental composition.

6. Results

We used the MCNP and ORIGEN2-A (Groff, 1997) codes for soil activation considering the elemental composition given by Soil A in Table 5. The model used fast (14 MeV) neutrons with emission rate of 10^9 n/s into 4π. The geometry considered was that of an isotropic point source placed 5 cm above the ground. The highest total induced activity after 10 h irradiation was 6.1×10^{-4} Ci, which decayed after one hour to 7.9×10^{-7} Ci and after one day to 8.6×10^{-8} Ci, which corresponded to about 17 pCi/g and initially consisted

Table 4
Main elemental content of organic components of soil organic matter

	C	H	O	N	S
Semi/cellulose and sugars	44.4	6.2	49.4		
Fatty acids	77.5	12.0	10.5		
Waxes	81.0	13.5	5.5		
Resin	80.0	11.5	9.0		
Enzymes	50.0	6.7	12.4	12.4	
Protein and amino acids	53.0	7.0	23.0	16.0	
Lignin and tannin	62.0	6.1	31.9		
Humic acids	56.2	4.7	35.5	3.2(0.8–5.5)	0.8(0.1–1.5)
Fulvic acids	45.7	5.4	44.8	2.1(0.9–3.3)	1.9(0.1–3.6)

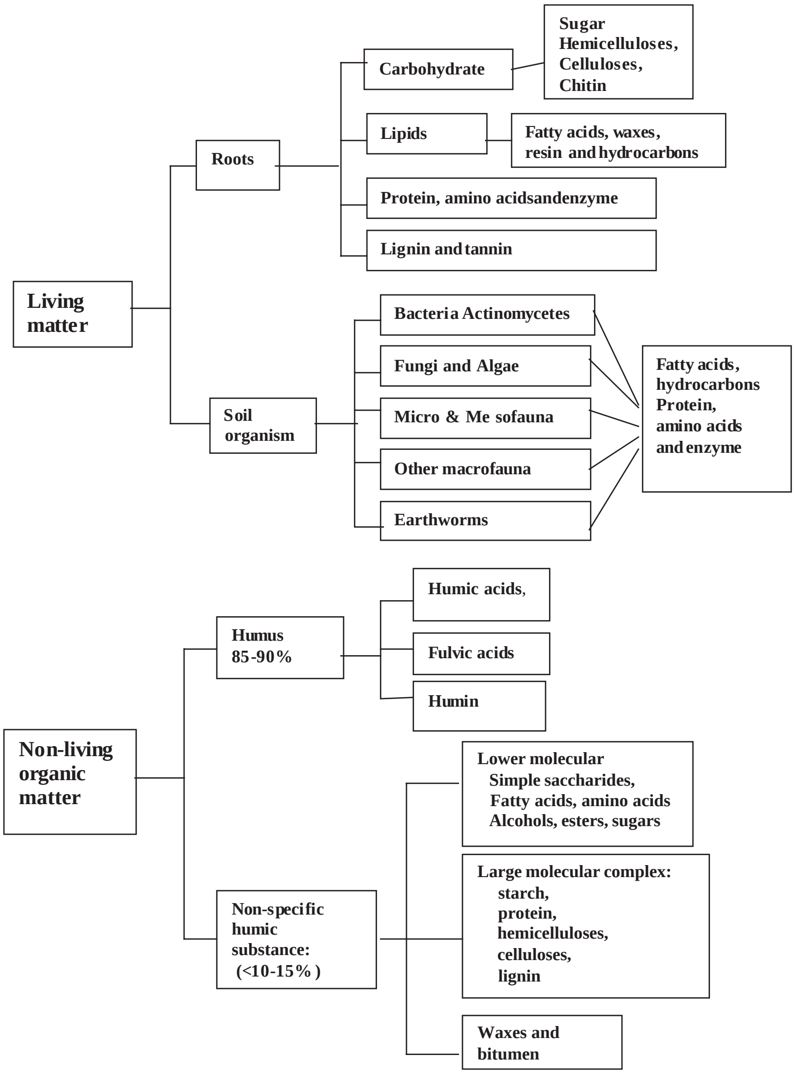

Fig. 5. Partial breakdown of the living and non-living organic matter.

Table 5
Elemental soil composition for; Soil A: Local soil chemical analysis at the laboratory, Soil B: Soil composition used in the data set of the PEGS4 code (Miller and Turk, 1951), and Soil C: is the median value of world soils, The Nanking Institute of Soil Science, 1982

Element	Soil A (% by weight)	Soil B (% by weight)	Soil C (% by weight)
^{1}H	0.2100	2.81	—
^{3}Li	0.0013	—	0.0030
^{5}B	0.0013	—	0.0010
^{6}C	0.1950	14.43	2.0000
^{7}N	0.0032	0.001	0.1000
^{8}O	53.0200	49.64	48.8700
^{9}F	0.0046	—	0.0200
^{11}Na	0.2450	0.82	0.6300
^{12}Mg	0.0881	—	0.5000
^{13}Al	1.0960	8.93	7.1000
^{14}Si	43.7600	21.32	33.3000
^{15}P	0.0128	—	0.0650
^{16}Cl	0.0108	—	0.0100
^{19}K	0.2250	0.56	1.4000
^{20}Ca	0.1180	0.54	1.3700
^{21}Sc	0.0005	—	0.0007
^{22}Ti	0.1228	—	0.5000
^{23}V	0.0035	—	0.0100
^{24}Cr	0.0029	—	0.1000
^{25}Mn	0.0151	—	0.0850
^{26}Fe	0.7240	0.96	3.8000
^{27}Co	0.0007	—	0.0008
^{28}Ni	0.0009	—	0.0040
^{29}Cu	0.0012	—	0.0020
^{37}Rb	0.0058	—	0.0100
^{39}Y	0.0035	—	0.0050
^{40}Zr	0.0545	—	0.0300
^{56}Ba	0.0540	—	0.0500
^{60}Nd	0.0063	—	—
^{62}Sm	0.0012	—	—
^{72}Hf	0.0029	—	0.0006
^{82}Pb	0.0050	—	0.0010
^{83}Bi	0.0051	—	—
^{90}Th	0.0037	—	0.0005

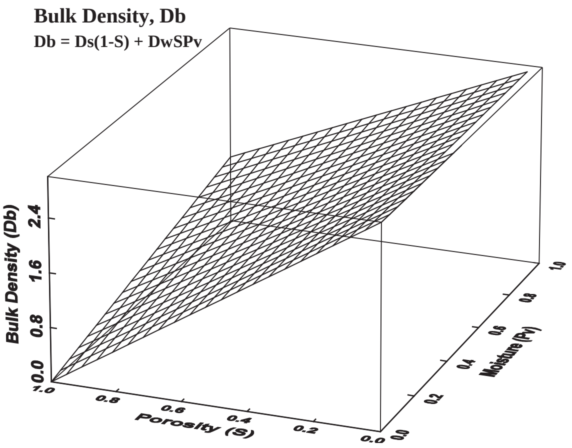

Fig. 6. Bulk density variation versus soil porosity and moisture fraction.

mainly due to activation of Al, Mg, N, Na and F, after 10 days the main activity was due to Ar and Sc. This might be compared to naturally occurring activity due primarily to thorium in soils at Long Island, New York, of 2.0×10^{-8} Ci. However, this activity was two orders of magnitude less than the natural activity of thorium.

Validity of these calculations and their sensitivity to soil composition were partially tested by comparing the neutron fluxes obtained when Soil B, in Table 5, was used versus results obtained when composition of Soil A was used. Furthermore, while Soil A used the mean neutron flux in a cell when using soil B the flux crossing the cell wall was calculated. All the other parameters were maintained the same. The neutron flux calculations of the high-energy group (5–14 MeV) as a function of depth for various water contents are shown in Fig. 7 for both soil compositions. The graph denoted as Alt. MCNP Calc in Fig. 7 was based on elemental

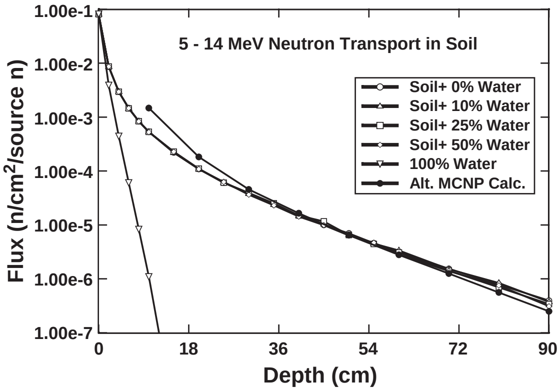

Fig. 7. The neutron flux intensity versus depth in soil for various soil water content. The solid squares represent calculation performed with different soil elemental composition.

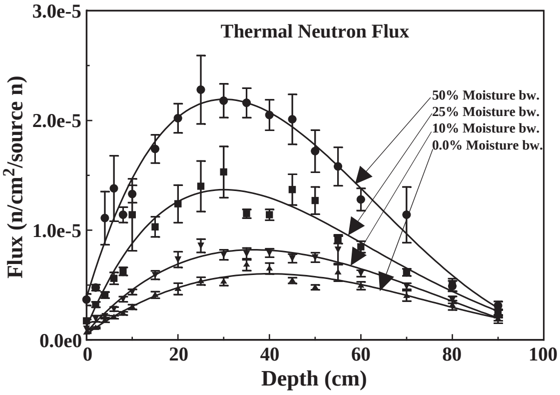

Fig. 8. Thermal neutrons flux for various soil moisture content.

composition of Soil A. Although, these neutron fluxes were calculated by two different and independent groups for different soil composition and neutron tallies, they are in general agreement. The large cell size in Alt. MCNP calculation prevented comparison of results below 10 cm. The main incentive for this manuscript pertains to gamma ray spectroscopy resulting from inelastic neutron scattering, which has threshold energy at about 4.8 MeV for carbon, thus the interest in neutron energies above that energy. Nevertheless, whenever the interest is in thermal neutrons the behavior of thermal neutrons as a function of soil moisture content is shown in Fig. 8. It is interesting to observe that the maximum flux occurs at about 40 cm deep and shifts to lower depth with increase in the soil moisture content. At 50% moisture it is about 30 cm. Thus optimization of the thermal neutron flux at shallower depth would require some pre-moderation. At 100% water the maximum occurs at about 3 cm.

Finally, the affect of soil bulk density on the neutron transport in the soil of the fast energy group was calculated for densities varying between 0.5–2.0 g/cm^3. The results are shown in Fig. 8.

7. Summary

In spite of the large variability in the soil parameters Monte Carlo calculations are widely used in radiation transport calculations in soil. However, lack of specific information on soil elemental composition, density, and density profile that varies with depth in the near surface layers hinders these calculations. It was pointed out that the significance of the various parameters on the analysis actually depends on the objectives of the calculations. For example, in the near surface layers, 10–15 cm, we showed in Fig. 9 that the variation in the soil density has very small effect on the neutron flux. Similarly, we also showed in Fig. 7 that soil moisture, up to 50% by weight, has negligible effect on the transport of the fast neutron energy group. This is mainly due to reduced elastic scattering cross section of hydrogen at neutron energies above 1 MeV. However, in Fig 8 the thermal neutron intensity almost doubles at $D_{\max}$, the peaking depth of the thermal neutrons, and there is a shift in $D_{\max}$ from about 40 cm to about 30 cm, which is due to increased thermalization. The thermal neutrons are of interest when considering prompt and delayed gamma ray spectroscopy following thermal neutron capture, whereas carbon and oxygen analysis is concern with fast neutron capture and the resulting inelastic neutron scattering. It was pointed out that depending on the objectives of the Monte Carlo calculation a less refined information might be required. For example, we examined the sensitivity of the fast neutron transport in soil for two neutron group and found no dependence

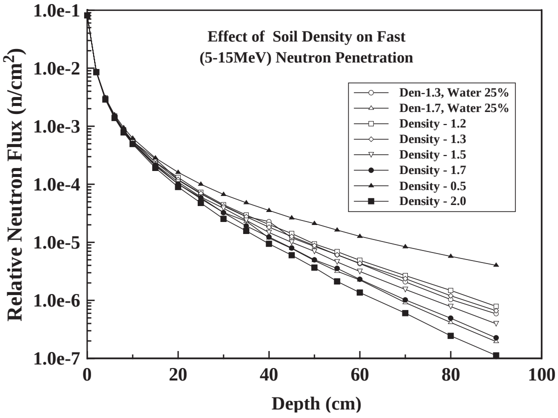

Fig. 9. Fast neutron flux distribution as a function of depth in soil for various soil densities.

on water content, Fig. 7, and weak dependence on density at larger depth. Indicating that precise information is not very critical. On the other hand when soil activation is of concern detailed elemental information of the trace level elements is required, as it was shown for short term and long term soil activation calculations.

An approximate road map has been presented to arrive for the soil elemental composition at a given site. Alternatively, if it is critically important, it is possible to take a soil sample and analyze it for the elemental composition. However, it is important to keep in mind to ascertain how representative this soil sample is of the entire site. Calculations to understand how individual parameters affect neutron and gamma radiation transport in soil are in progress and will be reported.

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