Continuous versus pulse neutron induced gamma spectroscopy for soil carbon analysis

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HIGHLIGHTS

- Calibration of the neutron induced gamma analysis system must account for system background and the interference of other nuclei (mainly silicon-28) on the carbon peak at 4.43 MeV.
- Spectra measured at a height of 250 cm above the ground could be considered the NGA system background spectrum.
- The experimental cascade transition coefficient for silicon-28 (i.e. ratio of 4.50–1.78 MeV gamma ray intensities) agrees well with theoretical calculations.
- The NGA continuous working mode halved the measurement time compared to the pulse working mode while retaining the same degree of accuracy.

ARTICLE INFO

ABSTRACT

Neutron induced gamma spectra analysis (NGA) provides a means of measuring carbon in large soil volumes without destructive sampling. Calibration of the NGA system must account for system background and the interference of other nuclei on the carbon peak at 4.43 MeV. Accounting for these factors produced measurements in agreement with theoretical considerations. The continuous NGA mode was twice as fast and just as accurate as the pulse mode, thus this mode was preferable for routine soil carbon analysis.

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1. Introduction

An emerging method for soil carbon analysis in the field is the measurement of gamma rays from soils subjected to neutron irradiation. There are different processes of neutron interaction with nuclei that produce gamma rays; measurement of this irradiation can be used to determine soil elements. For example, the inelastic neutron scattering (INS) of fast neutrons ($\sim$14 MeV) from carbon-12 nuclei produce gamma rays (4.43 MeV) that can be used to determine soil carbon (Wielopolski, 2011). Neutron induced gamma spectra analysis (NGA) has several advantages over other methods of carbon determination such as dry combustion, mid and near infrared reflectance spectroscopy, and laser induced breakdown spectroscopy. The NGA method is a non-destructive method that requires no sample preparation and is capable of analyzing large volumes of soil at once (Wielopolski, 2011). But determining the number of counts from gamma rays with energy of 4.43 MeV attributed to carbon is challenging using this method since several other nuclei and processes contribute to this peak in the gamma spectra. For example, some gamma rays that overlap this area are generated from neutron interactions with silicon-28 nuclei (Wielopolski, 2011; Basunia, 2013); silicon is one of the main components of soil. Other nuclei (i.e. aluminum-27, oxygen-16) under neutron irradiation can also contribute to this range of interest.

Neutron generation and gamma spectra acquisition can be performed in two modes: continuous and pulse. In the continuous mode, a single gamma spectrum encompassing INS processes, thermal neutron capture (TNC), delay neutron activation, and natural background is acquired. In the pulse mode, separate gamma spectra are acquired during and between neutron pulses. The gamma spectrum acquired during the pulse is similar to the continuous mode spectrum. The gamma spectrum acquired between pulses is attributed to TNC, delay neutron activation, and

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natural background; it is free from INS gamma rays. Gamma spectra acquisition during and between neutron pulses has an advantage over the continuous regime since it allows for more elaborate analysis of different elements and processes that are responsible for peaks in the spectra. However, in this mode the neutron irradiation intensity during the pulse depends on the duty cycle and frequency of the neutron generator setup (total number of neutron in unit time is constant) which is higher than the continuous mode. The disadvantages of the pulse mode occur at higher neutron fluxes where the background level is higher (increased pile-up overlapping in range of interest by low energy gammas), areas of peaks of interest are less (due to increasing dead time), so the signal-to-noise ratio in the range of interest is less, and relative error is higher.

An in-depth evaluation of measurement modes and subsequent data processing procedures for efficiently and accurately surveying soil carbon will be discussed. The mode most suitable for routine field measurement of soil carbon will be identified.

2. Materials and methods

The NGA measurement system for soil carbon analysis consists of a neutron generator, gamma detector, protective shielding, and construction materials (Wielopolski, 2011). A neutron generator (Model MP320; ThermoScientific, Waltham, MA) suitable for this method produces fast neutrons with energy of 14 MeV due to the nuclear reaction of tritium and deuterium. A neutron flux equal to $\sim 10^8$ neutron s^{-1} can be produced by this generator in the continuous or pulse modes with a frequency of 1–25 kHz and a duty cycle of 5–100% (100% duty cycle corresponds to the continuous or direct current (DC) mode of measurement). The cross section of inelastic neutron scattering on carbon-12 nuclei, leading to the production of 4.43 MeV gamma rays, is relatively large (0.21 barns for 14 MeV neutrons; MacFarlane, 2014). One or several NaI (Tl) detectors with individual operating volumes of ~ 2500 cm^3 were used for gamma ray detection. For data acquisition, split electronics with corresponding ProSpect v0.1.11-vega software (XIA LLC, Hayward, CA) allowed for acquiring either a single spectrum (continuous mode) or two spectra during and between pulses simultaneously (pulse mode). IGOR software was used for gamma-spectra analysis (WaveMetrics, 2013). It should be noted that the NaI(Tl) detector volume is more than adequate for full adsorption of gamma rays untill energy of 8–10 MeV (Saint-Gobain Crystals, 2009). The first approximation of adsorption efficiency for such a large crystal (12.5 cm × 12.5 cm × 15.2 cm) practically does not change with energy. According to Monte-Carlo calculations (Saint-Gobain Crystals, 2009), the "peak-to-total" ratio in the main range of interest (from 1.8 to 4.5 MeV) for the large crystal does not change significantly. For these reasons, the spectra correction based on the dependence of the efficiency of gamma-ray registration with energy (product of adsorption efficiency on the "peak-to-total" ratio) was not considered in this research.

Since soil carbon concentration is typically quite low, the useful signal (net peak area with centroid at 4.43 MeV) is only slightly higher than background (Wielopolski et al., 2008a); therefore, to accurately determine such a low useful signal, identifying all components in the peaks of interest (inclusive of background spectra) and implementing correct data processing is very important.

A critical step is to determine the NGA system background which is the spectrum radiated by the construction material under neutron irradiation. System background is always present and can be determined by raising the NGA system until the signal is unaffected by the ground or other large objects. In this study, the NGA system equipped with one gamma-detector was raised to

various heights over a sand substrate to determine at what height the sand substrate no longer affected the measured spectra.

A Monte Carlo simulation was undertaken to better understand the behavior of neutron simulated gamma spectra with changing height. A sodium iodide detector having a similar geometry to our measurement system (i.e., detector size, disposition of neutron source and detector, and size of aluminum shield between neutron source and detector) was used in conjunction with a Geant4 toolkit (Agostinelli et al., 2003). The gamma spectra were compared to gamma lines simulated by the Geant4 toolkit and G4NDL4.0 neutron database to identify the nuclei responsible for peaks of interest (Geant4, 2014).

Another critical step is to determine the portion of the 4.43 MeV peak contributed by silicon-28 interference. The cascade transition coefficient value that accounts for this interference was calculated from spectra measured from silicon samples with different layer thicknesses. The silicon samples (bulk density 1.26 g cm^{-3}) were prepared using granulated silicon placed in stainless steel boxes (40 cm × 40 cm × H where H varied from 1 to 20 cm). Boxed samples were attached to the NGA system and raised to a height of 260 cm prior to measurement.

Understanding the dependence of the carbon peak area in the neutron stimulated gamma-spectra versus the carbon concentration in the sample is important for calibration purposes. For NGA system calibration, mixtures of sand with granulated carbon were prepared, and subsequent measurements were carried out. The mixtures were prepared by mechanical mixing (manually or concrete mixer) sand with a specified amount of granulated coconut shells (~ 1.5 mm diameter; 100% carbon content) until uniformity was attained. Six samples with 1, 2.5, 5, 7.5, 10 and 15% carbon by weight (Cw%) were prepared. The bulk density of the mixture (d_{mix}, g cm^{-3}) with changing Cw% was described as follows:

$$d_{mix} = \frac{1.7 \times 0.52 \times 100}{Cw\% 1.7 + (100 - Cw\%) 0.52} \tag{1}$$

where 1.7 is bulk density of sand, g cm^{-3}; 0.52 is bulk density of coconut shells, g cm^{-3}.

Each sample mixture was placed in a stainless steel box (40 cm × 40 cm × 20 cm) and was attached to the NGA system; both were raised to a height of 260 cm prior to measurement.

Soil was also collected from an experimental soil bin located at the National Soil Dynamics Laboratory in Auburn, AL (Batchelor, 1984) to test the accuracy of the NGA measurements in both regimes. The selected soil was a Houston clay (very-fine, smectitic, thermic Oxyaquic Hapluderts) which had a relatively high carbon concentration (i.e., ~ 2.8). A sample was prepared by mixing ~ 5 w % additional granulated carbon for measurement via the NGA system. In addition, subsamples were collected from soil and sample mixtures, ground with a roller grinder (Kelley, 1994), and analyzed by dry combustion method utilizing a LECO TruSpec CN analyzer (LECO Corp., Saint Joseph, MI) for comparison.

3. Results and discussion

3.1. Measurement of system background

The background or portion of the spectra radiated by construction material was determined. Accounting for this background is critical since the carbon signal is only slightly higher than background (Wielopolski et al., 2008a) and thus is important for correcting all data. In this study, the NGA system equipped with one gamma-detector was raised to increasing heights over a sand substrate to determine at what height the signal was unaffected by the ground surface or other large objects.