PHAROS: A Pluri-detector, High-resolution, Analyser of Radiometric properties Of Soil

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Abstract

PHAROS is a new type of core logger, designed to measure activity concentrations of ⁴⁰K, ²³⁸U, ²³²Th and ¹³⁷Cs in sediment and rock cores with a spatial resolution of a few centimetres along the core. PHAROS has been developed as a non-destructive alternative to the traditional slicing of cores into sub-samples and their analysis on an HPGe detector. The core is scanned at fixed increments by three BGO scintillation detectors and the spectra analysed by the full spectrum analysis method. The core logger is also equipped with a collimated lead castle and a ¹³⁷Cs source for transmission measurements. In this presentation, we report on the properties of the core logger and its detectors, and on the analysis techniques used for the determination of the radionuclides activity concentrations. Results from initial measurements are presented and discussed.

1 Introduction

Drill cores of rock and sediment substrates play a significant role in many Earth Sciences disciplines as well as in the oil and gas exploration and mining industries. These cores are analysed to extract information on porosity, grain-size and type of sediment, which is obtained from a combination of physical quantities such as P-wave velocity, magnetic susceptibility, electric resistivity, gamma-ray attenuation and (natural) radioactivity. One of the crucial points in such analyses concerns the spatial resolution of the measuring apparatus and partly related to it, the measuring time to obtain statistically accurate data.

In climate research for instance, the knowledge of sedimentation rates and of the provenance of sediments present on the ocean floor is essential to reconstruct changes in ocean currents in relation to climate changes [1]. In these cases sedimentation rates are of the order of 1 mm per year and the information is needed on a decade level. Therefore, a spatial resolution of 1 cm or better is required to successfully assess the sedimentation rates on the ocean floor.

The PHAROS core logger (Pluri-detector, High-resolution, Analyser of Radiometric properties Of Soil) has been developed on request of the Earth Sciences community. It measures the natural

and anthropogenic radioactivity in soil and sediment drill cores with a high spatial resolution to provide accurate and detailed lithologic information. The gamma-ray activity of rocks and sediments is primarily given by the gamma decay of the naturally occurring radionuclide ⁴⁰K and of the decay products of the ²³⁸U and ²³²Th decay series. Moreover, anthropogenic radionuclides, which include ¹³⁷Cs, can be detected in recent sediments (>1950). These radionuclides result from episodic events like the atmospheric bomb-tests (1953-1963), the Chernobyl reactor accident (1986) or the incineration of radioactive sources (e.g. Algacieras, Spain in 1998), or come from the continuous release by the nuclear and chemical industries of waste materials in rivers and seas.

Commercially available core loggers [2] are mainly used for P-wave velocity, magnetic susceptibility and gamma-ray attenuation measurements, but seldom to measure activity concentrations of naturally occurring radionuclides. For high-resolution (1 to 2 cm) information on the ⁴⁰K, ²³⁸U and ²³²Th activity concentrations, the cores have to be sliced and each slice measured on hyper-pure germanium (HPGe) detectors. For the low-level activities in natural sediments the counting time per slice becomes of the order of a day. This makes analysing cores a tedious and consequently a costly procedure. One of the solutions is to use more efficient detection systems in a core logger. Such a detector system (MEDUSA) has been developed by the Nuclear Geophysics Division of the KVI (NGD) for surveys of the seafloor and in airborne surveys [3-5]. In this system a highly efficient detector material (bismuth germanate or BGO) is used in combination with a sophisticated analysis of the gamma-ray spectra. Compared to an equal size NaI and windows analysis, MEDUSA is an order of magnitude more sensitive [6]. In this paper we report on the properties of the core logger PHAROS and present some of the initial results.

2 Experimental set-up

Figure 1 shows a schematic vertical cross-section of PHAROS. A vertically mounted core (maximum length of 1 m) is kept in position by a hoisting system. A stepper motor is used to move the spindle and lower the core through the detectors and lead shielding at fixed increments. The stepper motor/spindle system is equipped with an absolute position encoder, allowing the position to be changed in mm steps. Moving a core up and down leads to a position uncertainty of 0.001mm. The control of the motor (VEXTA CSK564AE-TG30) and encoder is achieved via an I/O National Instruments Digital Acquisition (DAQ) PCI-6023E computer card and a circuit board made at the KVI. The I/O card comprises 8 digital ports and 2 pulse-generators/counters. The circuit board assures a good flow of signal to and from the motor and the encoder. Two lines are needed to control the motor, one for the direction of the rotation of the motor and the other one for when the rotation is needed.

A 16-input multiplexing multi-channel buffer (MCB) from EG&G Ortec is used for data acquisition. The MBC is connected to the acquisition PC via an Ethernet BnC network connection, allowing access from any PC on the same network.

In-house software was developed to process the core measurements automatically. The currently implemented software is essentially a batch process that handles several consecutive tasks repeatedly until any particular core measurement is complete.

At present, on the PHAROS upper-platform, three $Bi_4Ge_3O_{12}$ (BGO) detectors are mounted inside a 10 cm thick low-activity lead shielding (about 4000kg). The crystal's dimensions are 5 cm by 15 cm for all detectors. The detectors view the core through a 15 cm wide, and 10 cm deep slit. The height of the slit is 2 cm and can be adjusted in 5 mm steps. The detectors are positioned such that they each view a separate slice of the core, with 2 cm difference in height. In the present configuration 6 cm of the core are measured simultaneously. This way of scanning the core was chosen because of the very different background and spectral response of the three BGOs. This prohibits the addition of spectra and consequently necessitates a separate analysis of each spectrum. Due to the relatively high background of the BGO, the quality of the analysis improves with the measuring time. By treating the detectors separately and letting them scan adjacent parts of the core simultaneously, we obtained a better result than having all detectors viewing the same slice of core and reducing the measuring time linearly with the number of detectors. Opposite the third BGO detector (not represented on figure 1) a collimated lead castle has been mounted to place a ¹³⁷Cs source. This source is used for transmission measurements to determine the density of the core as a function of position. To avoid interference of the source with the radioactivity of the core, the density scans are carried out separately. The density measurements are primarily used to correct the activity concentrations for sediment density in the core but also to detect any void in the core, which could lead to erroneous interpretations of the results. PHAROS has additional platforms to install future equipment such as magnetic susceptibility and electric resistivity sensors.

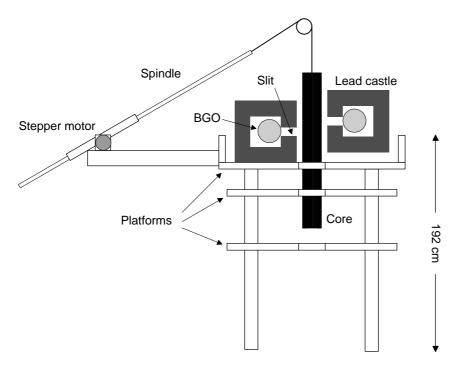


Figure 1: Schematic vertical cross-section of PHAROS. Only two detectors are represented on the drawing for clarity.

3 Analytical techniques

Gamma-ray spectra have been analysed using the full spectrum analysis method [6] to obtain activity concentrations of ⁴⁰K, ²³⁸U, ²³²Th and ¹³⁷Cs. Traditionally these concentrations are derived by setting windows in the gamma-ray spectrum at 0.66 MeV ¹³⁷Cs line, the 1.46 MeV ⁴⁰K line, the 1.77 MeV line of ²¹⁴Bi (U-series) and the 2.61 MeV line of ²⁰⁸Tl (Th series). The

deduction of the concentrations involves background subtraction and so-called stripping factors to account for Compton contributions of higher energetic gamma rays to the particular window [7]. Especially in this case where background dominates the gamma-ray spectrum and varies with time and relatively low activity concentrations are dealt with, the uncertainties become prohibitive.

As shown by Hendriks *et* al. [6] the combination of BGO and full spectrum analysis (FSA) leads to more than an order of magnitude larger sensitivity. Here we present the principles of the methods and refer for details to [4]. The FSA utilises all reliable information of the gamma-ray spectrum and uses a background spectrum *BG* and standard spectra P_j to reproduce the shape and intensity of the spectrum *S*. Standard spectra P_j represent the response of the detector to an activity concentration of 1 Bq/kg of radionuclide *j*. In FSA the measured spectrum *S* is described as the sum of the standard spectra P_j multiplied by the (unknown) activity

concentrations C_j for the individual radionuclide j plus a background spectrum adjustable in intensity. The activity concentrations follow from a fit of the calculated spectrum to the measured one and a least-squares procedure is used to find the optimal activity concentrations:

$$\boldsymbol{c}^{2} = \frac{1}{N - M - 1} \sum_{i=1}^{N} \left[S_{i} - \sum_{j}^{M - 1} C_{j} P_{ij} - \boldsymbol{a} B G_{i} \right]^{2} / w_{i}$$
(3.1)

In equation 3.1 *i* represents the channel number up to *N*, *M* is the number of standard spectra and w_i is a weighing factor set to be $w_i = \frac{1}{\mathbf{s}^2(S_i)}$ (3.2)

In the PHAROS case where the background varies with time, the shape of the background is kept constant but by treating it as a standard spectrum, its intensity a is allowed to be optimised. Consequently M also includes the background spectrum.

4 Results and discussion

Soil erosion and its subsequent redistribution is a major concern around the world. The use of environmental radionuclides, in particular ¹³⁷Cs, provides an effective method to make actual measurements of average soil loss and re-deposition quickly and efficiently. Traditional procedures for applying ¹³⁷Cs measurements in floodplain-sedimentation investigations involve the collection of sediment or soil cores from a study area and their subsequent transfer to the laboratory for slicing and analysis of soil samples on ¹³⁷Cs activity concentration by gamma spectrometry.

In cases where a large number of cores are collected, these procedures require substantial effort and long counting times (24 h per sample), as environmental samples usually contain low levels of radioactivity.

In its initial phase, PHAROS was intended mainly to provide a fast and accurate assessment of sedimentation rates based on the ¹³⁷Cs dating technique, although additional information on ⁴⁰K, ²³⁸U and ²³²Th is available. In its present stage, however, this additional information is exploited to detect alteration zones, the presence of heavy minerals and determine the type of sediment via a fingerprinting technique [3].

As a demonstration of the instrument and methods applied in PHAROS, we show the first ¹³⁷Cs profile of a 1 m long core (consisting of two 50 cm pieces) obtained during the commissioning of the core logger. The test core was drilled in August 2000 in the floodplain of the Meuse River near Itteren, the Netherlands. The core was measured in steps of 2 cm; each step measured for 3 hours, with an effective spatial resolution of 3 cm. The data were corrected for differences in density from the standard. The activity concentration as function of depth is presented in figure 2. The uncertainties in the data points reflect the uncertainties due to the counting statistics and the full spectrum analysis. The uncertainties are almost independent of the Cs activity concentration, indicating that the uncertainties are dominated by the background. Two peaks are observed in the profile, interpreted as being representative of the atmospheric bomb test around 1960 and the fallout of the Chernobyl reactor accident in 1986. The centroid of each peak was determined by fitting a Lorentzian to it. This procedure stems from the modelling of the migration of radionuclides in soil. Studies have shown that the depth distribution of Cs in undisturbed soils, several years after deposition, can be closely approximated by a 3-parameter Lorentz function [8]. This function characterises the observed distribution in all critical sections of the vertical profile: surface layer, distribution around the maximal concentration and the tail at greater depth.

The results of the fits give the positions at 20 and 60 cm. These positions correspond to average sedimentation rates of 1.4 ± 0.1 cm/year for the period 1986-2000 and 1.5 ± 0.1 cm/year for the period 1960-1986. Based on this result one may conclude that the average sedimentation rate has been quite constant for the entire period 1960-2000.

To assess the reliability of PHAROS, a parallel core of 50 cm was drilled about 30 cm besides the location of the measured one. The parallel core was sliced in 3 cm slices and the subsequent samples freeze-dried. Each sample was then measured on an HPGe detector for 24 hours. The results have been plotted together with the PHAROS data in figure 2. Both sets of data (PHAROS and HPGe) are in good agreement with each other, in particular for the position of the maximum of activity concentration and for the amplitude of the peak.

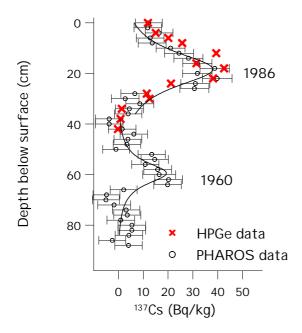


Figure 2: ¹³⁷Cs profile as a function of depth for a core drilled in the Meuse floodplain near Itteren. Both HPGe and PHAROS data are plotted. The continuous line represents the Lorentz fit to the PHAROS profile.

5 Conclusions and prospects

At the time of completion of this paper the effective spatial resolution PHAROS is 3 cm, which is sufficient to accurately assess modern sedimentation rates (>1960). The reliability of the PHAROS results has been tested by slicing some of the measured cores and by measuring the slices on an HPGe. The results obtained with PHAROS correspond well with those obtained from the HPGe analysis of the slices.

Campaigns in the Dutch floodplains of Itteren and Borgharen have been successfully carried out using PHAROS for the determination of sedimentation rates. Twenty-one meters of cores have been measured in 44 days. The same investigation carried out with an HPGe detector would have lasted more than a year.

Except for calibration and maintenance periods, PHAROS has been in constant use since August 2000 and proved to be an ideal instrument for the dating of sediment cores using ¹³⁷Cs as a time marker.

Our aim is to reach sub-centimetre resolution with PHAROS, but also to increase the measuring speed without deteriorating the quality of the results. The latter can be achieved by increasing the number of detectors on PHAROS to measure a higher number of adjacent parts of a core simultaneously.

The main obstacle preventing us from reaching a sub-centimetre resolution is the high background of the detectors caused by the impurities in the crystal, radiation from the PMT and the cosmic radiation reaching the BGO through the lead shielding. The first step in reducing the background is to use low background BGO crystals and potassium-free PMTs. The next stage involves the reduction of the contribution of the cosmic radiation to the background. We are presently investigating the possibility of mounting plastic scintillators on top of the lead shielding surrounding the BGOs to perform anti-coincidence measurements and mainly veto the contribution from muons to the final spectra. Deconvolution of the observed profiles into true profiles and apparatus broadening function is going to be addressed via MCNP simulations.

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