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Application of self-absorption correction method in gamma spectroscopy for ^{210}Pb and ^{137}Cs sediment chronology on the continental slope off NW Africa

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Abstract. The uppermost 50 cm of the high resolution core GeoB9501-4 from the continental slope off NW Africa has been used for improving routine technique of ^{210}Pb and ^{137}Cs sediment chronology in the Radioactivity measurement laboratory of the Bremen University. The experimental requirements for ^{210}Pb and ^{137}Cs determination were non-destructive analysis of wet sediment samples in cylindrical containers with different heights, unknown elemental composition and density. The method used in this study to deal with self attenuation is applying efficiencies calculated using the LabSOCS^{TM}, Genie 2000 software calibration tool, validated by self-absorption test measurements of different materials with known composition. A mean sedimentation rate of $0.49\,\mathrm{cm \cdot yr^{-1}}$ was obtained from the investigated core using the CRS chronological model. This means a relatively increased recent sedimentation rate comparing to an average sedimentation rate at higher depths of an associated gravity core estimated to $0.15\,\mathrm{cm \cdot yr^{-1}}$ by ^{14}C chronology. A further increase of the sedimentation rate was observed in the uppermost part of the studied core (up to $0.90\,\mathrm{cm \cdot yr^{-1}}$).

1. INTRODUCTION

1.1 Climatological background

Life in the semiarid Sahel belt of tropical North Africa strongly depends on the availability of water and has been repeatedly affected by shifts to more arid climate. The most recent drought occurred in the early 1970's and 1980's, with partial recovery during the late 1990's. High resolution fluvial sediments off Senegal offer the opportunity to study the history of the Sahel drought and to assess its imprint on the composition of terrestrial materials deposited at the sea floor, if the material can be accurately dated on historical time scales.

1.2 Sediment chronology

With the background of the upper mentioned project, we present ^{210}Pb and ^{137}Cs data from the high resolution multi-core GeoB9501-4 recovered during METEOR-Cruise M65/1 on the continental slope off NW Africa (Senegal Mudbelt, northern rim of Mauritanian Canyon, depth 330 m) – Fig. 1. The uppermost 50 cm of the multi-core has been used for improving the routine technique of ^{210}Pb and ^{137}Cs sediment chronology in the Radioactivity Measurement Laboratory of Bremen University.

1.3 Self attenuation

Since attenuation of emitted low-energy gamma radiation in voluminous bulk samples is an obstruction for determining ^{210}Pb (46.5 keV, Iγ 4.25%) quantitatively by means of gamma-spectroscopy, self-absorption correction must be taken into account. Two basic approaches have been applied for solving

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the problem of self-attenuation in volume samples: experimental [1, 2] and mathematical – using Monte Carlo simulations [3]. The approach combining both experimental measurements and mathematical MC simulations was proposed by other authors [4, 5].

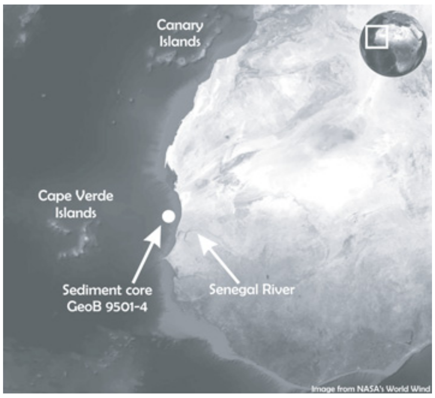


Figure 1. Map of the sampling point of the multi-core GeoB9501-4 recovered during METEOR-Cruise M65/1 on the continental slope off NW Africa (Senegal Mudbelt, northern rim of Mauritanian Canyon from the depth 330 m).

2. METHODS

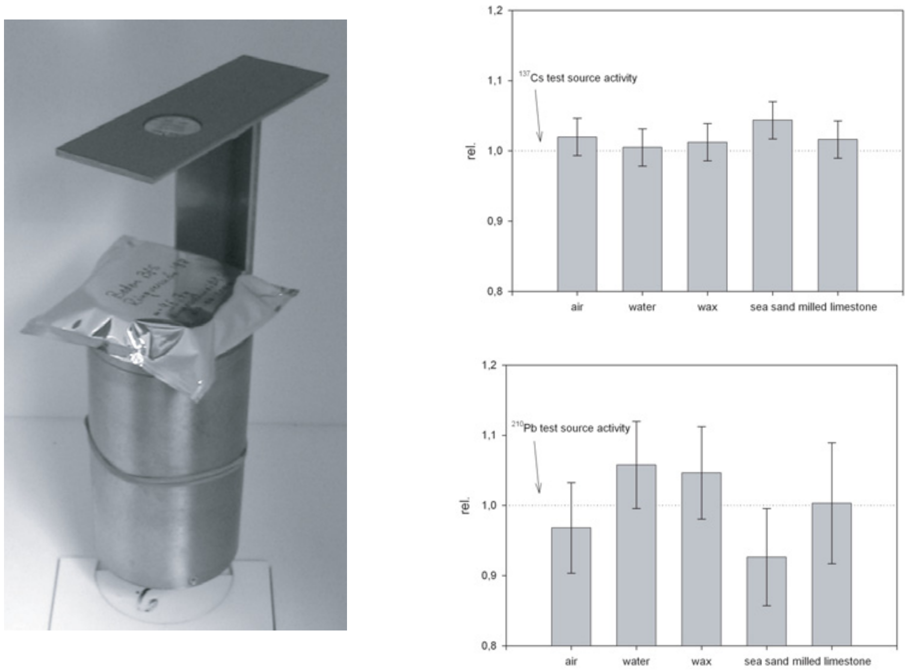
2.1 Gamma spectroscopy

A coaxial HPGe detector (Canberra Industries, 50% rel. efficiency) housed in a 10 cm Pb shielding with Cu and plastic lining operated under Genie 2000 software was used for low level, low-background gamma spectroscopy. The method used by the authors in this study to deal with self attenuation is applying efficiencies calculated using the LabSOCS^{TM} (Laboratory Sourceless Calibration System), Genie 2000 software calibration tool [6], validated by self-absorption measurements of different materials.

2.2 Material test

To validate the efficiency calibration generated by LabSOCS, a transmission experiment was realized for different absorbers. As emitters, point sources of gamma energies of 46.5 keV (^{210}Pb) and 661.6 keV (^{137}Cs) with reported activities (produced by Buchler) were used on a holder 15 cm above the absorber, which was placed directly on the detector (Fig. 2a). Five different materials with various chemical composition and densities were used as absorbers, all of them sealed in cylindrical plastic containers (round dishes) with diameter of 70 mm and height of 20 mm (the same containers were used for sediment samples): air (density $0\ g \cdot cm^{-3}$), water (density $1\ g \cdot cm^{-3}$), wax (composition: 15% H and 85% C, density $0.96\ g \cdot cm^{-3}$), sea sand (composition: 50% quartz, 50% K-feldspar, bulk density $1.4\ g \cdot cm^{-3}$) and milled limestone (composition: $CaCO_3$, bulk density $1.77\ g \cdot cm^{-3}$). The sample geometry modelling and efficiency calibration file generation were performed using the Geometry Composer feature of the Genie 2000 software. The efficiencies for given energies are estimated by LabSOCS for a characterized detector based on MCNP modelling code upon description of a sample container, absorber matrix, and a specific source-to-detector distance.

The efficiencies were used for analyzing gamma-spectra of the point sources using an experimental setup with the above listed absorbers and estimating their activities using the software Genie 2000. The gamma spectra of absorbers were collected separately and were subtracted from the spectra of the transmission experiments. The calculated activities of the point sources were compared to their reported activities (Fig. 2b). The experiment showed that activities estimated using LabSOCS generated efficiencies are within the overall error produced by Genie 2000.



a) b)

Figure 2. a) Transmission experiment setup showing the detector (bottom), a sealed sample (center) and transmission test source on its holder (top). b) Real relative activities of a ^{137}Cs (top) and a ^{210}Pb (bottom) point gamma test sources compared to activities estimated by measurement of their activities using absorbers of different chemical compositions and densities. The efficiencies were generated by LabSOCS.

2.3 Measurement of sediment samples

Wet sediment slices from each 1 cm interval of multi-core GeoB9501-4 were put into plastic round dishes (Petri dishes) with diameter of 70 mm and height of 20 mm directly after slicing of the core. It is advantageous to perform gamma spectroscopy before the core is processed due to the fact that the signal increases with the amount of material. Gamma spectroscopy as non-destructive method allows using the samples for other purposes after the analysis. The containers were filled with the samples to different heights varying from 5 to 20 mm depending on the available sample volume. For determination of $^{210}Pb_{exc.}$ activity, $^{210}Pb_{sup.}$ activity (determined via the 351.9 keV line of ^{214}Pb after establishing of Rn progeny equilibration) was subtracted from the $^{210}Pb_{total}$ signal. The spectra were analyzed using efficiencies generated by LabSOCS for different sample geometries, constant sediment composition (0.36% H, 2.14% C, 49.62% O, 0.84% Na, 1.60% Mg, 7.10% Al, 27.38% Si, 2.37% K, 4.21% Ca, 0.34% Ti and 4.04% Fe) and different densities varying from 1.0 to 1.7 g · cm^{-3}.

2.4 ^{210}Pb and ^{137}Cs dating

The age of the core has been estimated by ^{210}Pb chronology using the CRS model [8]. The model assumes a constant rate of supply of unsupported ^{210}Pb to the sediment per unit time and considers a variable sedimentation rate resulting from human activity or natural reasons, i.e., climate variability. Absolute ages were calculated with the assumption that the uppermost slice of the core corresponds to July 2005, the time of core retrieval. ^{137}Cs is present in the sediments due to the global fallout after nuclear bomb testing. It first appeared in the atmosphere in 1945 and peaked in 1963 at the northern hemisphere and can be therefore used for additional calibration of the age.

3. RESULTS AND CONCLUSIONS

Volume activities of $^{210}Pb_{exc.}$ *vs.* depth and volume activities of ^{137}Cs decay corrected to the relevant age *vs* age are shown in Fig. 3a.

According to the ANNEX C of the UNSCEAR report [8], the total amount of ^{137}Cs deposited from 1945 to 2000 in the latitudinal band of 10–20 degrees north recalculated to 2007 activities (time of the measurement) is $660\,Bq \cdot m^{-2}$. Comparing the total activity of $150\,Bq \cdot m^{-2}$ found in the core GeoB9501-4 to the UNSCEAR value, only 23% of the expected value was preserved in the sediment record. Due to rather low ^{137}Cs values the measurement errors are relatively high and do not provide fine resolution (expected 1963 peak or contribution of geographically close four Algerian atmospheric tests in 1960–61). Nevertheless, the shape of the ^{137}Cs profile is compatible with the bomb fallout chronology, possibly followed by a terrestrial (erosion-produced) component.

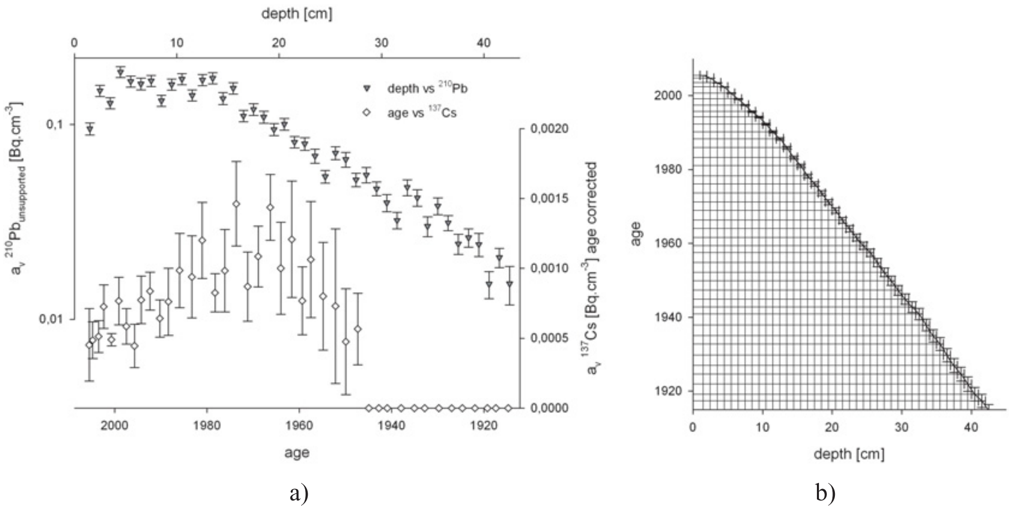


Figure 3. a) ^{210}Pb and ^{137}Cs data of gravity core GeoB9501-4. b) Sedimentation rate estimated from the $^{210}Pb_{exc.}$ using CRS chronology model.

A mean sedimentation rate of $0.49\,cm \cdot yr^{-1}$ was obtained from the investigated core with an increasing trend towards the present (Fig. 3b). From 1920's to 1980's the sedimentation rate is rather constant: $0.42\,cm \cdot yr^{-1}$, in 1990's the rate increases to an average $0.58\,cm \cdot yr^{-1}$ and in the 21st Century it reaches $0.90\,cm \cdot yr^{-1}$. Generally, a relatively increased recent sedimentation rate can be observed compared to an average sedimentation rate at older ages of the associated gravity core estimated to $0.15\,cm \cdot yr^{-1}$ by ^{14}C chronology (S. Mulitza, unpublished data).

Based on transmission test with different absorbers it can be concluded, that efficiencies produced by LabSOCS and used for activities estimations (including self-absorption correction) can thus be successfully used for purposes of combined ^{210}Pb and ^{137}Cs chronology.

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