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Intercomparison of alpha and gamma spectrometry techniques used in ^{210}Pb geochronology

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Abstract

^{210}Pb geochronology is a widely used tool in sedimentological studies aimed at the absolute ages of modern sediments (up to 100 years). ^{210}Pb activities required to model sedimentation regimes are measured using either alpha, gamma or beta spectrometry. Sediment accumulation rates derived from ^{210}Pb activity profiles measured by these methods are often used interchangeably in mass balance studies. Yet there is a lack of investigations considering the comparability of data derived using different analytical methods. Differences between methods could be caused by different behaviors of ^{210}Pb and ^{210}Po (used for alpha measurement) in the marine environment. In gamma spectrometry errors may arise when many gamma emitters are measured simultaneously and their activity peaks overlap. In alpha spectrometry chemical separation of ^{210}Po may result in analytical error due to incomplete sample dissolution.

In the present study we evaluate total, supported and excess ^{210}Pb activities and their use in deriving sediment accumulation rates and ^{210}Pb excess inventories for three sediment cores collected from the Barents Sea. ^{210}Pb activities derived by alpha and gamma methods are shown to agree within counting

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error and there is also good agreement in the derived sediment accumulation rates. The inherent compatibility of analytical results based on alpha or gamma techniques is established.
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1. Introduction

^{210}Pb geochronology is widely used to determine the ages of sediment layers in investigations of a variety of environmental processes. Common applications include assessments of material fluxes to the seafloor, environmental pollution studies, and inter-calibration of multiple tracers to determine event sequences in time (Carroll and Lerche, 2003; Robbins, 1978). The ^{210}Pb method, first introduced by Goldberg (1963), has been applied in studies of lakes, estuaries and coastal marine sediments. ^{210}Pb (half life = 22.3 years) is a member of the uranium-238 decay series. In shallow ocean areas, it is supplied by the *in situ* disintegration of ^{222}Rn ($t_{1/2} = 3.8$ days) from dissolved ^{226}Ra in seawater and from atmospheric deposition of ^{210}Pb to sea surfaces. Lead, including ^{210}Pb, is particle reactive, and thereby readily sorbs to sinking particles in the ocean (Cochran, 1982). Because bottom sediments also contain ^{210}Pb from the *in situ* decay of radium-226 (supported ^{210}Pb), the additional ^{210}Pb is known as excess or unsupported ^{210}Pb ($^{210}Pb_{ex}$). The activity of $^{210}Pb_{ex}$ decreases over time since deposition while $^{210}Pb_{supp}$ activity remains constant.

A ^{210}Pb-chronology is determined for a sediment core based on the down-core activities of $^{210}Pb_{ex}$ (total minus supported). There are three alternative methods of analysing the total activity of ^{210}Pb in a sediment sample:

- beta spectrometry: measurement of ^{210}Bi activity,
- gamma spectrometry (herein called gamma): direct ^{210}Pb measurement (^{210}Pb emits a 46.5 keV gamma photon),
- alpha spectrometry (herein called alpha): measurement of ^{210}Po activity.

Supported ^{210}Pb ($^{210}Pb_{supp}$) by gamma spectrometry is achieved by measuring the activity of other daughter products of ^{226}Ra (i.e. ^{214}Bi, ^{214}Pb) simultaneously with total ^{210}Pb on each sediment depth interval. In the case of alpha spectrometry, $^{210}Pb_{supp}$ is determined by measurement of ^{210}Pb sufficiently deep below the sediment–water interface where no $^{210}Pb_{ex}$ remains and a single value is applied to all sediment depth intervals. In all cases, $^{210}Pb_{ex}$ is estimated as the difference between total and $^{210}Pb_{supp}$ activity.

The use of alpha measurements in ^{210}Pb dating assumes that ^{210}Po–^{210}Pb is in equilibrium so that by measuring ^{210}Po activity we also derive ^{210}Pb activity. This is not always the case. ^{210}Po is more reactive with organic matter than ^{210}Pb (Cochran, 1982). So in some cases, there is ^{210}Po enrichment in fresh, surface sediment layers. To avoid this problem samples are stored for several ^{210}Po half-lives (138 days).

Profiles of $^{210}Pb_{ex}$ versus depth (cm) and mass depth (g/cm^2) are then used to calculate sediment accumulation rates and sedimentation rates, respectively (Carroll and Lerche, 2003). The Constant Initial Concentration (CIC) model is appropriate when initial activity of $^{210}Pb_{ex}$ is constant and there is no mixing of surface sediments (Robbins and Edgington,

1975). This implies deposition of sedimentary material characterized by constant $^{210}Pb_{ex}$ activity such that either both $^{210}Pb_{ex}$ activity and mass of deposited material to the sediment surface are constant or both vary at the same rate. The Constant Rate of Supply (CRS) model is used when the supply of $^{210}Pb_{ex}$ is constant and the sediment deposition rate is variable (Robbins, 1978; Appleby and Oldfield, 1978). There are also more advanced procedures available in cases where both sediment accumulation rates and the supply of $^{210}Pb_{ex}$ vary with time (Carroll et al., 1999; Carroll and Lerche, 2003).

When a $^{210}Pb_{ex}$ profile is disturbed due to surface mixing, sediment accumulation rate is usually assessed for the portion of the profile below the mixed layer.

Regardless of which model is chosen, sediment accumulation rates depend on the shape of the $^{210}Pb_{ex}$ profile ($^{210}Pb_{ex}$ activity versus depth corrected for sediment compaction), and on the individual results of the $^{210}Pb_{ex}$ activity measurements.

Often sediment accumulation rates derived from $^{210}Pb_{ex}$ activity profiles measured by the alpha and gamma methods are used interchangeably in environmental studies (Smith et al., 2002). Hence it is necessary to perform a systematic comparison of the precision and accuracy of activity results and sediment accumulation rates obtained using both analytical methods. Tanner et al. (2000) also conducted a comparison study of alpha and gamma methods but only on measurements of a few depth intervals from cores collected in Hong Kong Harbour. They report a systematic error with larger gamma measured ^{210}Pb activities but concluded that alpha and gamma methods are comparable.

Here we conduct a more detailed investigation, comparing ^{210}Pb measurements (total, supported and unsupported) carried out by gamma and alpha spectrometry techniques and applying the determined radionuclide activity profiles to derive sediment accumulation rates and $^{210}Pb_{ex}$ inventories. The study was carried out on sediment cores from three stations in the Barents Sea where sediment accumulation rates are on the order of 1 mm/year.

There are few studies concerning sediments and sedimentation processes in the Barents Sea. The access to this area is limited and sampling is difficult. The present work is part of a large research project on sedimentary processes related to carbon flux in the Barents Sea. The stations selected for these methods' intercomparison are influenced by diverse physical and biological controls: permanent or seasonally ice covered waters, warm and cold currents, and different primary productivity regimes. We postulate that the results of the present work are broadly applicable in other continental shelf seas.

2. Materials and methods

2.1. Field methods

Sediment cores from three stations along a south–north transect in the Barents Sea were collected in 2003 (stations III and IV) and 2004 (station XII) (Table 1) using a multicorer with four, 50 cm long core and 10 cm diameter tubes. Three sediment cores from each cast were directed for gamma measurements while the fourth core was used for alpha measurements. Cores for gamma analyses were sliced every 5 mm (0–5 cm intervals) and 10 mm (10–20 cm intervals). Similar sediment depth intervals from each core were combined, dried at 60 °C and ground into fine particles. The fourth sediment core for alpha was sliced every 10 mm and frozen until analysis. Sediment sub-samples were removed and placed into pre-calibrated vials for porosity determinations. Sediment sub-samples were also retained for analyses of grain size and organic carbon content.

Table 1
Sampling station description, range of porosity, grain size and organic carbon content for stations III, IV and XII

Station	Latitude °N	Longitude °E	Depth (m)	Core length (cm)	Porosity	Grain size – pelite fraction (%)	Organic carbon content (%C)
III	79° 01′	25° 46′	198	22	0.82–0.64	97–89	1.4–1.2
IV	77° 01′	29° 29′	222	28	0.77–0.45	54–45	1.4–1.0
XII	80° 09′	29° 36′	286	20	0.85–0.66	95–86	1.5–1.3

2.2. Laboratory methods

2.2.1. Gamma spectrometry

Twenty grams of dried sediment was placed in counting vials and stored in plastic containers for 21 days to allow complete ^{222}Rn decay. Gamma emitting radionuclides were measured in ORTEC, high-purity, planar germanium detectors at ENEA (The Italian National Agency for New Technologies, Energy and the Environment). Total ^{210}Pb was determined by measurement of the 46.5 keV gamma peak; $^{210}Pb_{supp}$ (^{226}Ra) was determined by measurement of ^{214}Pb (at 295 and 352 keV) and ^{214}Bi (at 609 keV). The $^{210}Pb_{ex}$ activity was determined by subtracting $^{210}Pb_{supp}$ (average of ^{214}Pb and ^{214}Bi activities) from total ^{210}Pb for each depth interval. Detector efficiencies were calibrated using several sources, and confirmed using IAEA standard material (IAEA-300). Blanks were equal to natural background, assessed from measurements of an empty detector taken over several days. Sediment activities were corrected for self-absorption (Cutshall et al., 1983).

2.2.2. Alpha spectrometry

Frozen samples for alpha analysis were transported to Institute of Oceanology, Polish Academy of Sciences (IOPAS) where they were dried at 60 °C and ground. Radiochemical separation of ^{210}Po was performed by the method developed by Flynn (1968) and adopted by Pempkowiak (1991). Sediment samples of 0.2 g were spiked with ^{209}Po and digested using perchloric acid and hydrofluoric acid. Polonium isotopes were spontaneously deposited in acid solution (ion exchange reaction) onto silver disks. After deposition, disks were washed with methanol and analysed for ^{210}Po and ^{209}Po in a multi-channel analyzer (Canberra) equipped with Si/Li detectors. The samples were counted for 1 day. The activity of ^{210}Po in the sample was determined based on chemical recovery by comparing the measured and spiked activities of ^{209}Po. Efficiency of detection was calculated for every sample (30–36%). Blanks and standards (IAEA-326) were measured to verify the efficiency of the separation procedure and detection. The $^{210}Pb_{supp}$ activity was calculated as the average of several ^{210}Pb determinations in deep sediment layers (below the zone of ^{210}Pb exponential decline).

2.2.3. Sediment accumulation rates and inventories

$^{210}Pb_{ex}$ versus depth profiles were corrected for time of collection and compaction based on measurements of sediment porosity (calculation was done using sample porosity and dry density). Sediment accumulation rates were calculated using both the CIC and CRS models. In the CIC model, the dependence between $^{210}Pb_{ex}$ activity after time t and $^{210}Pb_{ex}$ activity at the sediment surface is given by:

$$A_x = A_0\, e^{-\lambda x/v}$$

where A_x is the $^{210}Pb_{ex}$ activity at layer x (cm below sediment–water interface), A_0 is the $^{210}Pb_{ex}$ activity of the surface layer, λ is the ^{210}Pb decay constant (0.031/year) and v is the sediment accumulation rate (cm/year). A constant sediment accumulation rate is derived from

$$v = -\lambda/a$$

where a is the slope of the line derived from a linear regression analysis of the $\ln(^{210}Pb_{ex})$ versus depth profile.

In the CRS model:

$$A_x = A_c\, e^{-\lambda/t}$$

$$x/v = 1/\lambda\ \ln \sum A_{cx} / \ln \sum A_{c0}$$

where $\sum A_{cx}$ is the cumulative residual $^{210}Pb_{ex}$ activity beneath sediments of depth x, $\sum A_{c0}$ is the total $^{210}Pb_{ex}$ activity in the sediment column, v is the sediment accumulation rate (cm/year), x is the cumulative depth (cm), t is the age of sediment (years) and λ is the decay constant. Where sediment mixing of surface layers was indicated (stations IV and XII), the accumulation rate was determined by applying the CIC model below the mixed depth.

Inventories of $^{210}Pb_{ex}$ were calculated based on the cumulative sum of $^{210}Pb_{ex}$ (Bq/kg) determined by both alpha and gamma methods. These values were multiplied by the cumulative mass of each sediment layer (g/cm^2), and expressed in Bq/m^2.

3. Results

The sediment cores collected at three locations exhibit comparable porosity values and organic carbon content but different grain size compositions. The pelite fraction (silt and clay fractions) is lower at station IV compared to stations III and XII (Table 1). $^{210}Pb_{ex}$ profiles at stations IV and XII indicate surface mixing to approximately 3 cm depth (Table 1).

3.1. Total ^{210}Pb activities

Total ^{210}Pb activities in sediments are similar at stations III and IV (max. = 272 Bq/kg), but higher at station XII (max. = 422 Bq/kg) (Table 2). Station XII is the deepest station (Table 1) and therefore longer particle residence times in the water column allow more time for ^{210}Pb sorption prior to seafloor deposition. In general, total ^{210}Pb activities determined by gamma are higher than for alpha at stations III and IV while both gamma and alpha total ^{210}Pb activities are comparable at station XIII. These trends are confirmed by performing a linear regression of gamma versus alpha data from each core (Fig. 1a). Combining all cores together, there is a slight reduction from a 1:1 correspondence (slope = 0.95; $R^2 = 0.93$) due to the higher gamma activities observed in surface sediments at stations III and IV.

3.2. ^{210}Pb supported activities

$^{210}Pb_{supp}$ values for gamma are the mean of ^{214}Pb and ^{214}Bi in all measured layers and for alpha are the mean of layers with constant activity. $^{210}Pb_{supp}$ values are 55 Bq/kg (gamma) and 46 Bq/kg (alpha) for station III. A similar trend is observed at station IV with $^{210}Pb_{supp}$ by gamma equal to 30 Bq/kg and by alpha equal to 20 Bq/kg. However, at station III $^{210}Pb_{supp}$ activities by gamma showed a decrease with depth toward the alpha $^{210}Pb_{supp}$ estimate, while at station IV all gamma values were higher than alpha. At station XII, both the average gamma supported value and alpha value were identical at 41 Bq/kg.

Table 2
Activities of ^{210}Pb total, supported and excess (Bq/kg) in the cores III, IV and XII

Station	Layer (cm)	Total ^{210}Pb				$^{210}Pb_{supported}$				$^{210}Pb_{excess}$			
		Gamma	Error	Alpha	Error	Gamma	Error	Alpha	Error	Gamma	Error	Alpha	Error
III	0–0.5	261.1	14.1	218.5	5.0	63.8	5.4	46.5	3.5	197.2	19.8	171.9	13.5
III	0.5–1	242.2	23.5	182.4	5.6	62.2	2.9	46.5	3.5	180.0	19.3	135.9	11.0
III	1–2	210.8	27.2	166.4	5.4	61.8	4.2	46.5	3.5	149.0	21.7	119.9	9.8
III	2–3	159.6	18.4	140.8	5.0	58.0	4.1	46.5	3.5	101.5	13.7	94.3	7.8
III	3–4	138.5	16.5	89.3	3.9	64.8	4.2	46.5	3.5	73.8	10.0	42.8	3.7
III	4–5	105.4	8.8	80.9	3.8	55.5	3.7	46.5	3.5	49.9	5.4	34.4	3.1
III	5–6	79.4	6.0	63.7	3.3	51.5	2.8	46.5	3.5	27.9	2.6	17.2	1.6
III	6–7	69.4	4.2	67.0	3.4	52.8	2.7	46.5	3.5	16.6	1.3	20.5	1.9
III	7–8	60.6	2.7	62.7	3.3	52.3	2.7	46.5	3.5	8.3	0.6	16.1	1.5
III	8–9	48.8	2.4	41.8	2.7	55.3	2.7	46.5	3.5	−6.5	0.5	−4.8	0.5
III	9–10	52.9	2.9	46.7	2.9	55.3	2.7	46.5	3.5	−2.3	0.2	0.2	0.0
III	10–12	65.1	3.9	51.7	3.0	49.8	2.6	46.5	3.5	15.3	1.2	5.2	0.5
III	12–14	45.7	2.0			49.5	2.3			−3.7	0.2		
III	14–16	53.2	2.7	46.8	2.9	53.6	2.6	46.5	3.5	−0.4	0.0	0.3	0.0
III	16–18	57.1	2.3			50.0	3.8			7.2	0.6		
III	18–20	43.8	2.4			51.7	2.5			−7.9	0.6		
III	20–22			45.8	2.8			46.5	3.5			−0.7	0.1
IV	0–0.5	271.9	25.1	211.5	6.1	28.3	4.1	19.5	1.1	243.5	54.8	192.0	12.5
IV	0.5–1	242.5	23.5	202.8	5.9	34.2	3.8	19.5	1.1	208.2	55.7	183.3	12.0
IV	1–2	218.4	33.2	196.2	5.8	27.9	3.7	19.5	1.1	190.5	68.8	176.7	11.6
IV	2–3	209.6	28.2	190.9	5.8	30.9	3.8	19.5	1.1	178.7	63.6	171.5	11.3
IV	3–4	77.6	6.2	100.1	4.2	25.4	3.2	19.5	1.1	52.3	15.5	80.6	5.8
IV	4–5	44.4	3.8	64.4	3.3	30.8	3.0	19.5	1.1	13.6	0.5	44.9	3.5
IV	5–6	22.3	0.7	35.2	2.5	28.0	2.8	19.5	1.1	−5.7	3.4	15.7	1.4
IV	6–7	9.6	0.2	27.3	2.2	32.6	2.6	19.5	1.1	−22.9	9.1	7.8	0.5
IV	7–8	26.1	1.3	17.8	1.3	29.5	2.5	19.5	1.1	−3.4	5.5	−1.7	0.2
IV	8–9	11.2	0.6	20.4	1.3	32.1	2.5	19.5	1.1	−20.9	6.4	0.7	0.0
IV	9–10			19.1	1.9			19.5	1.1			−0.3	0.0
IV	10–12	11.3	0.4	20.7	1.9	28.3	2.4	19.5	1.1	−17.0	8.4	1.2	0.1
IV	12–14			18.1	1.7			19.5	1.1			−1.4	0.2
IV	14–16	19.3	0.7			33.5	2.3			−14.2	4.2		
IV	16–18			20.4	1.9			19.5	1.1			0.9	0.1
IV	18–20	37.8	2.0			25.9	2.4			11.9	0.9		
IV	20–22												
IV	22–24			18.4	1.8			19.5	1.1			−1.0	0.1
IV	24–26												
IV	26–28			21.2	1.9			19.5	1.1			1.7	0.2
XII	0–0.5	401.6	16.9	418.2	8.5	44.3	3.4	41.0	1.6	357.3	31.6	377.2	16.3
XII	0.5–1	409.6	16.7	383.2	8.3	44.1	4.2	41.0	1.6	365.4	38.0	334.9	14.6
XII	1–2	405.7	17.0	412.4	8.5	56.1	3.9	41.0	1.6	349.6	28.5	371.4	16.1
XII	2–3	288.7	13.0	362.8	7.9	46.1	3.6	41.0	1.6	242.6	21.8	321.8	14.0
XII	3–4	256.7	11.4	295.4	6.9	41.2	2.3	41.0	1.6	215.5	15.4	254.4	11.2
XII	4–5	228.2	9.0	259.6	6.7	42.0	2.3	41.0	1.6	186.2	12.6	218.6	9.8
XII	5–6			176.5	5.5			41.0	1.6			135.5	6.2
XII	6–7	83.1	7.1	131.6	4.8	36.2	1.8	41.0	1.6	46.9	4.6	90.6	4.2
XII	7–8	54.4	5.9	108.6	4.3	35.7	2.2	41.0	1.6	18.7	2.3	67.6	3.1
XII	8–9	57.9	6.7	75.0	3.6	42.2	2.4	41.0	1.6	15.7	2.0	34.0	1.5
XII	9–10			61.6	3.3			41.0	1.6			20.6	0.9
XII	10–12	40.1	5.0	42.8	2.7	35.0	2.1	41.0	1.6	5.1	0.7	1.8	0.1

(*continued on next page*)

Table 2 (*continued*)

Station	Layer (cm)	Total ^{210}Pb				^{210}Pb$_{supported}$				^{210}Pb$_{excess}$			
		Gamma	Error	Alpha	Error	Gamma	Error	Alpha	Error	Gamma	Error	Alpha	Error
XII	12–14			41.8	2.7			41.0	1.6			0.8	0.0
XII	14–16	55.2	6.9	38.9	2.6	36.9	2.2	41.0	1.6	18.3	2.5	−2.0	0.1
XII	16–18			39.7	2.6			41.0	1.6			−1.3	0.1
XII	18–20	48.7	3.0	41.8	2.7	34.7	1.3	41.0	1.6	14.0	1.0	0.8	0.0

3.3. ^{210}Pb excess activities

Trends in ^{210}Pb$_{ex}$ measured by alpha and gamma spectrometry are similar to those previously reported for total ^{210}Pb. Both station III and station IV exhibit higher ^{210}Pb$_{ex}$ by gamma compared to alpha. For example at station III, the mean difference in surface (0–5 cm) values is 40 Bq/kg while for deeper layers (5–20 cm) only 6 Bq/kg. At station IV, the average difference is 31 Bq/kg in surface intervals (0–5 cm) and 12 Bq/kg in deeper layers (5–20 cm). In the case of station XII, ^{210}Pb$_{ex}$ in surface sediments determined by alpha are actually higher than those measured by gamma. The mean differences are equal to 37 Bq/kg in surface layers (0–7 cm) and 10 Bq/kg in deeper intervals (7–20 cm). However, linear regression analysis of gamma versus alpha activities indicates that the differences are less than those previously observed in total ^{210}Pb (Fig. 1b). Here the slope of the regression line is 1.00 ($R^2 = 0.95$).

3.4. ^{210}Pb excess inventories

The ^{210}Pb$_{ex}$ inventories integrate differences in the measured activities determined by alpha and gamma methods over the entire profile (Fig. 2). For all stations, the inventories are comparable. For station III ^{210}Pb$_{ex}$ inventories derived by alpha and gamma are 0.26 and 0.32 Bq/cm^2, respectively. Slightly higher but similar ^{210}Pb$_{ex}$ inventories are reported for station IV with 0.48 Bq/cm^2 by alpha and 0.45 Bq/cm^2 by gamma. Inventories at station XII were higher than at the other stations, again due to longer particle residence times in the water column before burial compared to the other stations. The ^{210}Pb$_{ex}$ inventories were 0.94 and 0.94 Bq/cm^2 by alpha and gamma, respectively.

3.5. Sediment accumulation rates

We compare sediment accumulation rates derived for all stations using both the CIC and CRS models (Table 3). Station III sediment accumulation rates derived by the CIC method using alpha measurements equal 1.3 mm/year, while for gamma the rate is slightly lower at 1.1 mm/year. Similar rates are derived by the CRS method (Table 3).

At station IV, strong surface sediment mixing is observed with equal ^{210}Pb$_{ex}$ activities in the uppermost three layers. Sediment accumulation rates are therefore based on the exponential profile below the surface mixed layer depth. Once again, the rates using both analytical methods and models are comparable, ranging from 0.3 to 0.4 mm/year (Table 3).

Finally, for station XII, two surface intervals contain equal ^{210}Pb$_{ex}$ activities as a result of sediment mixing. In all cases the rates are similar both when considering different analytical

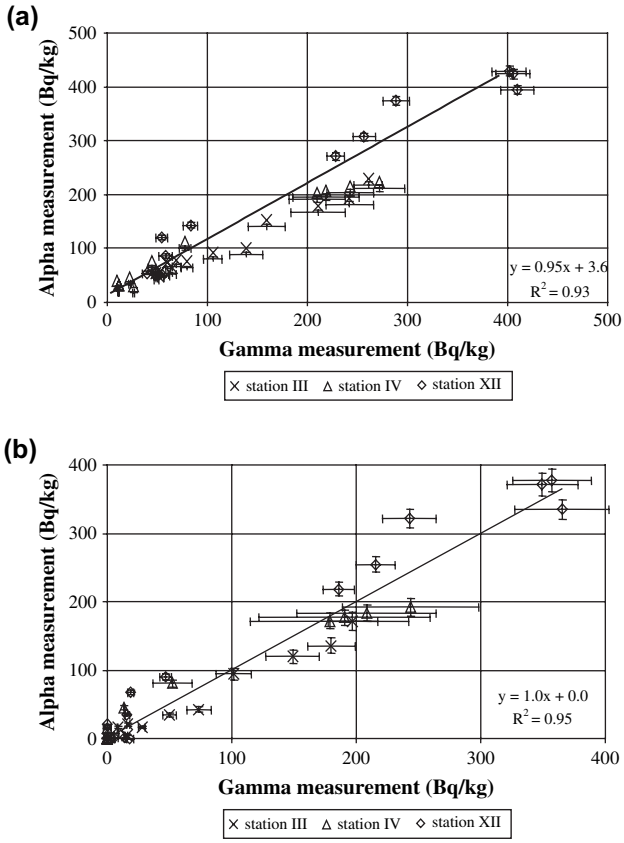

Fig. 1. (a) Total ^{210}Pb and (b) $^{210}Pb_{ex}$ activities (Bq/kg) derived by alpha versus gamma measurement techniques.

techniques and when applying different model procedures. Sediment accumulation rates below the mixed depth range from 1.0 to 1.1 mm/year (Table 3).

4. Discussion

Through this detailed comparison of analytical methods used in the determination of sediment accumulation rates by ^{210}Pb geochronology, we find that both alpha and gamma analysis techniques provide comparable results. The ^{210}Pb measurements, both supported and excess, agree within counting error. We do observe a trend of higher activities by gamma in the upper depth intervals at two of the three stations but the errors connected to these measurements are also quite large. For example, we report that at stations III and IV, surface intervals are on average 49 and 35 Bq/kg higher by gamma compared to alpha. If calibration standards are part of the cause, then it must be an activity dependent error at higher activities indicating, the derived efficiency coefficients, probably for gamma, are not as reliable. As shown in Fig. 2, the effect is largely eliminated after subtracting the supported from total ^{210}Pb activities to derive $^{210}Pb_{ex}$. A similar systematic error between measuring techniques was also noticed by Tanner et al. (2000). However, our station XII is an exception where

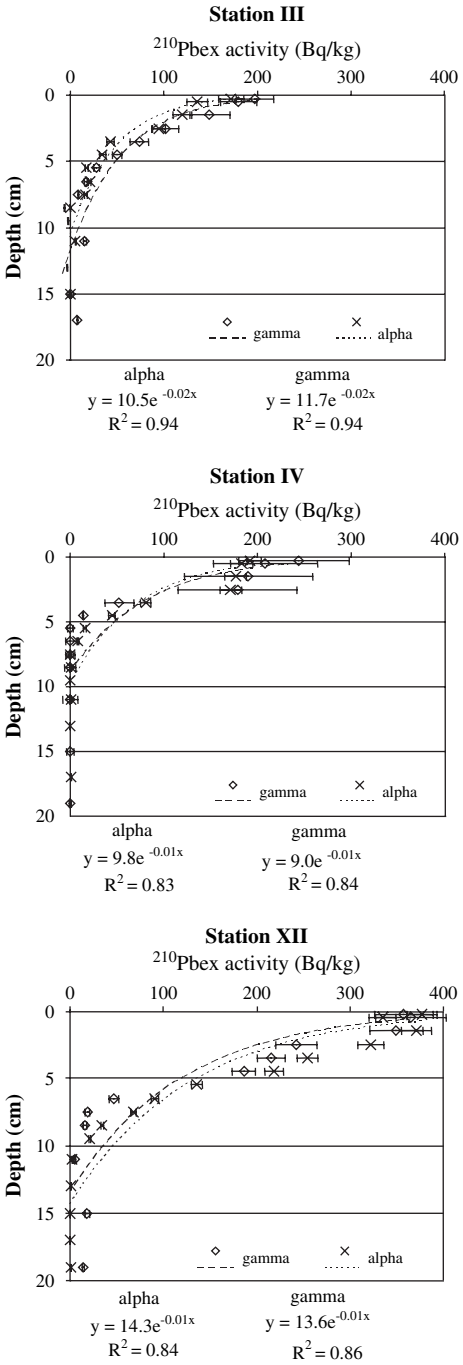

Fig. 2. ^{210}Pb excess activities (Bq/kg) versus depth for all stations.

Table 3
Sediment accumulation rates based on two methods ($^{210}Pb_{ex}$ by alpha, $^{210}Pb_{ex}$ by gamma) in addition to, approach with results of sediment accumulation rates from $^{210}Pb_{ex}$ alpha–gamma ($^{210}Pb_{total}$ by alpha $-^{210}Pb_{supp.}$ by gamma) is added

Stations	Sediment accumulation rate (mm/year)											
	Alpha method				Gamma method				Alpha–gamma method			
	CIC	R^2	CRS	R^2	CIC	R^2	CRS	R^2	CIC	R^2	CRS	R^2
III	1.3	0.93	1.1	0.91	1.1	0.99	1.0	0.97	1.1	0.91	1.0	0.89
IV	0.4	0.99	0.4	0.98	0.3	0.99	0.3	0.99	0.3	0.97	0.3	0.95
XII	1.1	0.99	1.0	0.95	1.1	0.92	1.0	0.82	1.3	0.89	1.1	0.78

For determination of sedimentation rates two models, CIC and CRS, are used.

activities of total, supported, and excess ^{210}Pb agree quite well. Stations are located in diverse depositional environments, influenced by different water masses, and different primary production regimes with different organic matter quantity and quality. Bottom topography of station locations and lateral sediment transport may also have influenced our reported $^{210}Pb_{ex}$ and $^{210}Pb_{supp}$ activities.

Some of the observed differences between gamma and alpha might be explained by environmental factors such as patchiness of sediment properties among cores or even within a single core. Within a single core, changes in $^{210}Pb_{supp}$ activity are often related to variations in particle composition such as grain size, organic carbon content (Smith and Walton, 1980; Chanton et al., 1989). For example station IV was characterized by variable and high sand content. This may lead to inaccuracies in supported and total ^{210}Pb values by alpha since $^{210}Pb_{supp}$ values derived from gamma are determined for each depth interval and thus already account for within-core variations such as in grain size.

Insufficient precision in slicing and combining sediment intervals for analysis by the gamma technique is another possible explanation for observed differences. However, differences in $^{210}Pb_{ex}$ resulting from application of the different methods are minimized through subtraction of $^{210}Pb_{supp}$ from total ^{210}Pb as both gamma data series (supported and total) contain slightly higher activities compared to alpha (stations III and IV only). In general, however, because the errors associated with $^{210}Pb_{ex}$ activity are high due to the determination of $^{210}Pb_{ex}$ through subtraction of two measurements (total ^{210}Pb and $^{210}Pb_{supp}$), $^{210}Pb_{ex}$ derived from both alpha and gamma analytical methods are within error bars for the measurements.

It is quite common in geochronology studies to combine data acquired by both alpha and gamma analytical methods to establish profiles of $^{210}Pb_{ex}$ activity. In some studies, total ^{210}Pb is determined by the alpha technique while $^{210}Pb_{supp}$ is acquired using the gamma technique. We also tested this approach on our data set. We derived sediment accumulation rates from $^{210}Pb_{ex}$ activity (alpha-gamma), applying both the CIC and CRS models. Sediment accumulation rates by the CIC and CRS models, determined by the combined (alpha-gamma) approach, are not only comparable to one another but also to rates determined using the gamma and alpha data sets (Table 3).

Hence both the rates and $^{210}Pb_{ex}$ inventories based on both gamma and alpha profiles are comparable even when using different models to derive sediment accumulation rates (Table 3). Mixing of surface layers at both stations IV and XII had little influence on the results. It is clear that both alpha and gamma methods are equally suitable for use in sedimentation investigations. Regardless of which method is chosen, the results can be compared with confidence.

4.1. Advantages and disadvantages of methods

The alpha method is superior in precision and is more suitable if only small samples are available. In this study, we have used just 0.2 g of dry sediment. Since ^{210}Po is measured after separation from the matrix there is no "background" to be subtracted. Alpha radioactivity detectors are also less expensive, and therefore more widely available in different laboratories. One of the disadvantages is the need to indirectly establish the $^{210}Pb_{supp}$ activity and the possibility of ^{210}Po–^{210}Pb disequilibrium. ^{210}Po is more reactive with organic matter than ^{210}Pb. In studies performed by Kharkar et al. (1976) on plankton, a very high ^{210}Po excess activity compared to ^{210}Pb was found. However, this is rarely observed for sediments, only in plankton or water samples (Gaboury and Hemond, 1988). But in some cases, there may be ^{210}Po enrichment in fresh, surface sediment layers. If the enrichment is very high even after several half-lives of sample storage before measurement, not all ^{210}Po excess would be lost through radioactive decay.

On the other hand, the alpha method is characterized by a time consuming separation process. Chemical separation of ^{210}Po can result in analytical error. While it is assumed that the standard procedure (Flynn, 1968) removes all ^{210}Po from solution, some authors report high separation efficiencies (80–99%) (Garcia-Orellana and Garcia-Leon, 2002). If chemical separation of ^{210}Po is not complete the analytical error will be higher. This is controlled through the addition of an internal standard. Our laboratory experience is that ^{210}Po deposition efficiency is $99 \pm 3\%$ ($n = 31$).

The alpha method is based on indirect ^{210}Pb measurement (^{210}Po). Hence there is an additional assumption of isotopic equilibrium between ^{210}Pb and ^{210}Po. In some cases surface sediments can be enriched in ^{210}Po because of its higher scavenging rate and hence the requisite equilibrium is lacking (Eakins and Morrison, 1978).

The gamma method allows for the non-destructive analysis of several gamma emitting radionuclides simultaneously in an individual sample. With no chemical separation procedure, the method is also easier and time efficient. However, with the exception of some high-purity well germanium detectors (Carroll et al., 1993), large sample sizes (20–50 g) and high measurement errors are associated with this technique. In some cases gamma measurement of ^{226}Ra may not reflect $^{210}Pb_{supp}$ due to the presence of additional sources of ^{226}Ra in the environment, thus leading to inaccurate estimation of $^{210}Pb_{supp}$. This situation was encountered by Theng et al. (2003), where ^{226}Ra activities were higher than total ^{210}Pb activities due to a riverine source of ^{226}Ra.

Using the gamma technique there is also the possibility of an error connected to self-absorption (Cutshall et al., 1983) and there is a requirement of time-consuming efficiency calibrations (Appleby, 2001). Gamma detectors are also more expensive, and require liquid nitrogen.

Surface sediment layers are also often mixed by physical processes or bioturbation (Robbins and Edgington, 1975). This mixing alters tracer distributions and, if not accounted for properly, leads to inaccurate interpretations of ^{210}Pb profiles (Smith, 2001). Therefore, independent tracers should be an essential component of sediment geochronology investigations in order to demonstrate consistency among results obtained by different tracers (e.g. ^{137}Cs).

5. Conclusions

Radionuclide activities based on both alpha and gamma analytical methods are comparable within measurement errors. With the exception of some high-purity well germanium

detectors, large sample sizes and high measurement errors for gamma analysis make alpha techniques attractive in some circumstances even if this method is time and labor consuming. Sediment accumulation rates and inventories derived from $^{210}Pb_{ex}$ profiles by both gamma and alpha techniques are shown to be similar making it appropriate to consider issues other than methodological inconsistency, when organizing a scientific investigation, to include ^{210}Pb geochronology.

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