

On the sediment age estimated by ^{210}Pb dating: probably misleading “prolonging” and multiple-factor-caused “loss”

JIA Jianjun¹, YANG Yang^{2*}, CAI Tinglu³, GAO Jianhua², XIA Xiaoming³, LI Yan⁴, GAO Shu¹

¹ State Key Laboratory of Estuarine and Coastal Research, East China Normal University, Shanghai 200062, China

² Key Laboratory of Coast and Island Development of MOE, Nanjing University, Nanjing 210023, China

³ State Research Centre for Island Exploitation and Management, Second Institute of Oceanography, State Oceanic Administration, Hangzhou 310012, China

⁴ College of the Environment and Ecology, Xiamen University, Xiamen 361102, China

Received 5 January 2018; accepted 9 February 2018

© Chinese Society for Oceanography and Springer-Verlag GmbH Germany, part of Springer Nature 2018

Abstract

The radionuclide ^{210}Pb is suitable for century-scale dating and has been used to calculate the sedimentation rate in a variety of environments. However, two common ways to apply ^{210}Pb dating techniques may give misleading results. One is “prolonging of age”, i.e., using the calculated sedimentation rate to date back to 200 or 300 years. This practice must be treated with caution because the ^{210}Pb dating techniques do not guarantee direct dating for ages much older than 100 years. Another is “loss of age”, i.e., the calculated time span between the topmost layer and the ^{210}Pb background layer in cores is less than 100 years when an apparent sedimentation rate is used in the calculation. Here, we propose that based on the principle of ^{210}Pb dating, the upper limit of age suitable for direct ^{210}Pb dating is between 110 and 155 years. The “prolonging” application is acceptable only if the sedimentary environment in the past several hundred years was stable and the sedimentation rate was generally constant, and verification with independent evidence (such as historical records or biomarker methodology) is needed. Furthermore, after analyzing many published and collected data, we found four possible reasons for the “loss of age”. First, the compaction effect of sediment should be corrected in laboratory analysis or else the calculated age will be underestimated. Second, the accuracy and uncertainty of ^{210}Pb activity measurement affect the judgment of the background. To be cautious, researchers are apt to choose a background activity with a younger age. Third, use of a slightly smaller value of supported ^{210}Pb activity in a calculation will lead to considerable underestimation of the time span. Finally, later-stage erosion and migration are common for sedimentation, which lead to loss of sedimentary records and are often reflected as a “loss of age” in cores. We believe that proper use of ^{210}Pb dating data may provide helpful information on our understanding of sediment records and recent environmental changes.

Key words: ^{210}Pb dating, sedimentation rate, sediment flux

Citation: Jia Jianjun, Yang Yang, Cai Tinglu, Gao Jianhua, Xia Xiaoming, Li Yan, Gao Shu. 2018. On the sediment age estimated by ^{210}Pb dating: probably misleading “prolonging” and multiple-factor-caused “loss”. *Acta Oceanologica Sinica*, 37(6): 30–39, doi: 10.1007/s13131-018-1214-4

1 Introduction

The natural radionuclide ^{210}Pb , an intermediate daughter of the ^{238}U decay series, has a half-time of 22.26 years and is obtained from the 3α decay, 2β decay and 1α decay of ^{226}Ra (half-time of 1 602 years) (Fig. 1). The daughter of the first α decay is the noble gas element ^{222}Rn , whose half-time is only 3.82 d (Liu, 2010). The ^{210}Pb in sediments have two major sources, unsupported and supported. The unsupported ^{210}Pb primarily originated from atmospheric deposition (including wet and dry deposition, as the decay daughter of ^{222}Rn), and it is then absorbed by suspended particulates and eventually enters sediment. This fraction of ^{210}Pb is also called “excess ^{210}Pb ” and denoted $^{210}\text{Pb}_{\text{ex}}$. The supported ^{210}Pb is the decay daughter of ^{226}Ra in sediments and is denoted $^{210}\text{Pb}_{\text{eq}}$. Generally, if disturbance, erosion and diffusion in sediments are ignored, the $^{210}\text{Pb}_{\text{ex}}$ that enters sediments will no longer receive atmospheric supply and will follow the general decay law of radionuclides, i.e., its activity decreases by

half every 22.26 years. While the $^{210}\text{Pb}_{\text{eq}}$ and precursor ^{226}Ra follow the long-term equilibrium relation of radioactive decay, the change in $^{210}\text{Pb}_{\text{eq}}$ within 100 years is negligible. When the above conditions are satisfied, we can infer the age of the layers by measuring the radioactivity of $^{210}\text{Pb}_{\text{ex}}$ at different layers in short cores. Combining this result with the depth and density of layers, we can calculate the sedimentation rate (unit: $\text{g}/(\text{cm}^2\cdot\text{a})$) or apparent deposition rate (unit: cm/a).

Ever since Goldberg (1963) proposed the principle of ^{210}Pb dating, geologists have widely applied the principle to dating and sedimentation rate calculations for ice cores, soils, lakes, estuaries, tidal flats, lagoons, bays and inner shelves (Koide et al., 1972; Nittrouer et al., 1979; Zou et al., 1982; Appleby and Oldfield, 1983; Qian et al., 1985; DeMaster et al., 1985; Alexander et al., 1991; Li et al., 1996; Wan, 1997; Xia et al., 1999, 2004; Chen et al., 2004; Zhang et al., 2009; Liu et al., 2009; Jia et al., 2012). It has become a powerful tool for studying century-scale sedimentary records and

Foundation item: The National Natural Science Foundation of China under contract Nos 41376068 and 41776068.

*Corresponding author, E-mail: yangy@nju.edu.cn

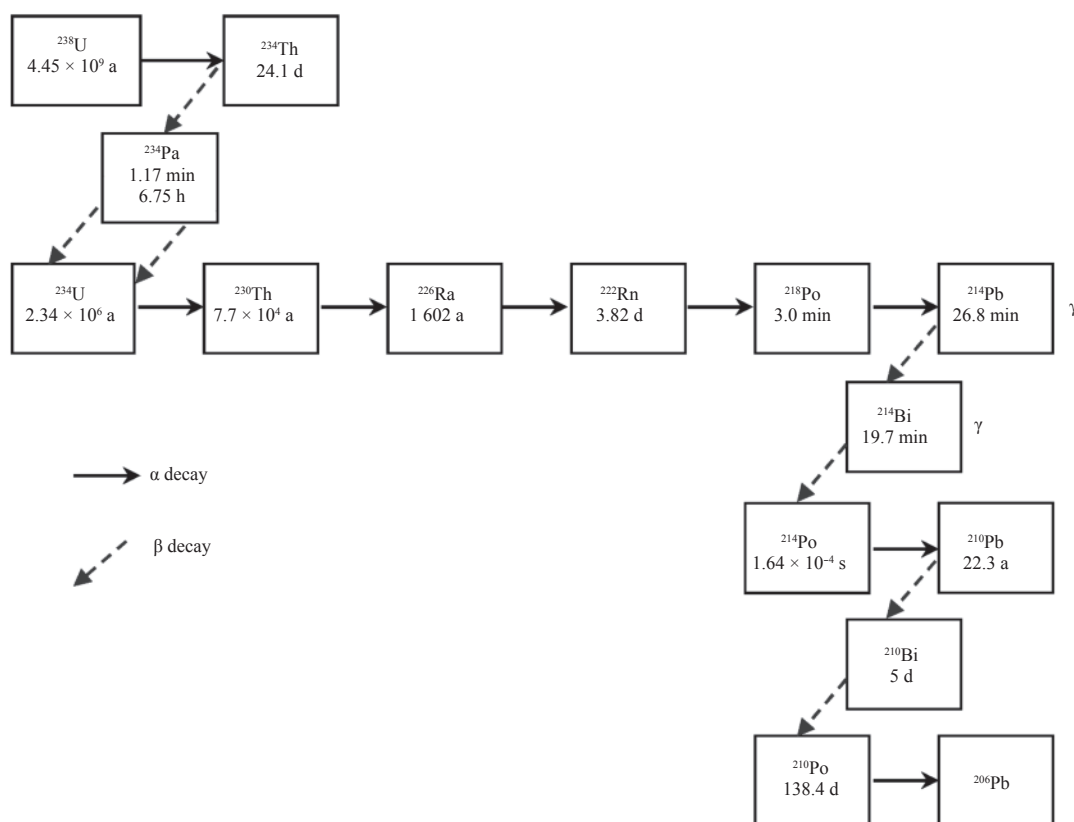


Fig. 1. U-Th decay series.

environment evolution. However, Binford (1990) once remarked, nearly 30 years after the technique of ^{210}Pb dating was proposed, that “ ^{210}Pb -dating are described mathematically in numerous papers, but actual calculation methods are never explicit. Estimates of dating uncertainty are seldom presented in published papers or reports”. Even today, this comment can still cause resonance among researchers.

Based on a literature review, we found two phenomena in the applications of ^{210}Pb dating techniques. One is “prolonging”, i.e., the dating age is backtracked using the calculated sedimentation rate; the backtracking period can reach 200 or 300 years in certain cases. The other is “loss of age”, i.e., the period above the ^{210}Pb background layer is calculated according to the measured apparent deposition rate and is less than 100 years in many cases. Both phenomena have certain problems. The application of “prolonging” already exceeds the time scale ensured by the long-term equilibrium between the precursor ^{226}Ra and daughter ^{210}Pb . The “loss of age” phenomenon may be due to several problems or errors that occur during laboratory pretreatment of samples, interpretation of data measured by an energy spectrometer or calculation procedures adopted for determination of ages, or it may indicate disturbances or missing layers in sedimentary records. However, for a long time, the former was applied thoughtlessly, and the latter was presented blindly.

Based on the principle of ^{210}Pb dating, the “prolonging” and “loss of age” phenomena are discussed in this study with collected ^{210}Pb data of short cores. We assessed the suitability and reliability of the former and examined the causes of the latter and precautions in interpretation. The objective of this paper is to promote a better understanding of ^{210}Pb dating data.

2 Principle of ^{210}Pb dating and sedimentation rate calculation

Two of the most common mathematical models for ^{210}Pb dating (Oldfield and Appleby, 1984) are the constant initial concentration (CIC) model and the constant rate of supply (CRS) model. More ^{210}Pb dating models are described by Wan (1997), Zhang et al. (2008) and Sanchez-Cabeza and Ruiz-Fernández (2012).

2.1 The CIC model

The basic assumption of the CIC model is that the suspended particles in water absorb $^{210}\text{Pb}_{\text{ex}}$ proportionally. When the suspended particle flux deposited into the water-sediment interface increases, the absorbed $^{210}\text{Pb}_{\text{ex}}$ flux that enters the water-sediment interface also increases. In other words, regardless of how the accumulation rate of sediment changes at the water-sediment interface, the $^{210}\text{Pb}_{\text{ex}}$ concentration at the water-sediment interface is constant. Therefore, the variation in the $^{210}\text{Pb}_{\text{ex}}$ concentration in the sediments with depth follows the following equation (Appleby and Oldfield, 1983):

$$C = C_0 e^{-\lambda t}, \quad (1)$$

where C_0 is the initial $^{210}\text{Pb}_{\text{ex}}$ activity (dpm/g) at the water-sediment interface, C is the $^{210}\text{Pb}_{\text{ex}}$ activity (dpm/g) at a layer in the sediments, λ is the ^{210}Pb decay constant ($3.114 \times 10^{-2} \text{ a}^{-1}$), and t is the layer age (a) corresponding to C , which can be calculated according to the following equation:

$$t = \frac{1}{\lambda} \ln \frac{C_0}{C}. \quad (2)$$

Sedimentary records suitable for the CIC model should satisfy

fy the following conditions (Appleby and Oldfield, 1983).

(1) The variation in the $^{210}\text{Pb}_{\text{ex}}$ concentration with increasing depth should be monotonic and always decreasing.

(2) The differences in the accumulative $^{210}\text{Pb}_{\text{ex}}$ flux among different cores from the same sedimentary environment (such as a lake) should be approximately proportional to the difference in the sedimentation rate.

2.2 CRS model

The basic assumption of the CRS model is that for certain local regions, the deposition flux of atmospheric $^{210}\text{Pb}_{\text{ex}}$ entering the water-sediment interface is constant. Because the suspended particles in water absorb $^{210}\text{Pb}_{\text{ex}}$ rapidly and efficiently, the deposition flux of the $^{210}\text{Pb}_{\text{ex}}$ in water entering the water-sediment interface is also constant and not affected by the sediment accumulation rate. Therefore, the relation between the accumulated $^{210}\text{Pb}_{\text{ex}}$ in sediments and age is expressed by the following equation (Appleby and Oldfield, 1983):

$$A_t = A_0 e^{-\lambda t}, \quad (3)$$

where A_0 is the accumulative activity of all the $^{210}\text{Pb}_{\text{ex}}$ from the water-sediment interface down to the background, and A_t is the accumulative activity of the $^{210}\text{Pb}_{\text{ex}}$ from a layer in the sediments down to the background. Both can be obtained by integrating the $^{210}\text{Pb}_{\text{ex}}$ activity of the corresponding intervals of cores. Additionally, t is the age of the layer corresponding to A_t and is calculated as follows:

$$t = \frac{1}{\lambda} \ln \frac{A_0}{A_t}. \quad (4)$$

Sedimentary records suitable for the CRS model should satisfy the following three conditions (Appleby and Oldfield, 1983).

(1) The variation curve of the $^{210}\text{Pb}_{\text{ex}}$ activity in sediments may show some fluctuation due to changes in the sediment accumulation rate because an increase in the sedimentation rate can decrease the initial $^{210}\text{Pb}_{\text{ex}}$ activity at the water-sediment interface and vice versa.

(2) For different cores taken from the same or similar sedimentary environments (such as from the same lake or certain regions with generally similar sedimentary environment characteristics), although their sedimentation rates may vary, the total deposition flux of $^{210}\text{Pb}_{\text{ex}}$ is generally similar.

(3) The total accumulated $^{210}\text{Pb}_{\text{ex}}$ in cores should reflect the atmospheric $^{210}\text{Pb}_{\text{ex}}$ deposition flux in the region.

The mature algorithm of the CRS model was first proposed in 1978 (Appleby and Oldfield, 1978; Robbins, 1978). When it was applied to lacustrine sediments dating, the results matched extremely well with the lamina record of lacustrine sediments. This work was published in *Nature* and drew broad attention (Appleby and Oldfield, 1979).

2.3 Sedimentation rate calculation

Suppose the sedimentation rate of sediments is R (g/(cm²·a)); then,

$$t = \frac{M}{R}, \quad (5)$$

where M is the mass depth (g/cm²) corresponding to age t (a).

If we use the CIC model to calculate the sedimentation rate, we substitute Eq. (5) in Eq. (1) and then take the logarithm, as fol-

lows:

$$\ln C = -M \frac{\lambda}{R} + \ln C_0. \quad (6)$$

Equation (6) is a one-dimensional linear equation of the form $y=ax+b$, where $y=\ln C$, $x=M$, $a = -\frac{\lambda}{R}$ and $b=\ln C_0$. We take the logarithm of the $^{210}\text{Pb}_{\text{ex}}$ of each layer in the cores and plot against the corresponding mass depth M ; then, the sedimentation rate is $R=-\lambda/b$.

If we use the CRS model to calculate the sedimentation rate, the deposition flux (R , g/(cm²·a)) in a certain layer can be obtained:

$$R = \frac{\lambda A_0}{A_t}, \quad (7)$$

Subsequently, the apparent sedimentation rate (r , cm/a) is expressed as follows:

$$r = \frac{R}{\rho}, \quad (8)$$

where ρ (g/cm³) is the bulk density of the sediment. For simplicity, several researchers have assumed that the bulk density of the sediment from the top to the bottom of a core is constant. The apparent sedimentation rate is intuitive; however, the assumption of constant bulk density of the sediment is an important reason for the “loss of age” phenomenon in ^{210}Pb dating. We will further analyze this below.

3 The upper limit of age and “prolonging” in ^{210}Pb dating

3.1 The upper limit of age for ^{210}Pb dating

According to the principle introduced in Section 2, the $^{210}\text{Pb}_{\text{ex}}$ variation with time is the key for ^{210}Pb dating and the calculation of sedimentation rates. Thus, we need to know the value of $^{210}\text{Pb}_{\text{eq}}$ (i.e., the ^{210}Pb background). There are generally two procedures to obtaining $^{210}\text{Pb}_{\text{eq}}$ (Su et al., 1984). For Procedure I, ^{226}Ra and ^{210}Pb in sediments are assumed to have already reached equilibrium. Thus, we take the ^{226}Ra radioactivity as a constant within a century-scale and then directly measure the precursor ^{226}Ra activity as the background, i.e., $^{210}\text{Pb}_{\text{eq}}$. For Procedure II, we observe the $^{210}\text{Pb}_{\text{total}}$ variation with depth and generally find a stable value of $^{210}\text{Pb}_{\text{total}}$ below a certain depth. We take this stable value as $^{210}\text{Pb}_{\text{eq}}$.

Fundamentally, both procedures rely on the long-term equilibrium principle of a radioactive decay series. According to this principle, if the half-time of the precursor is extremely long and if the half-lives of all daughter products are relatively short, the entire decay series reaches long-term equilibrium after a sufficiently long time. A decay series that has reached long-term equilibrium has an important characteristic, i.e., the activities of the precursor nuclide and daughter nuclides are equal, and all nuclides decay following the decay law of the precursor. Therefore, the abovementioned two procedures for determining the ^{210}Pb background measure either the precursor activity or the daughter activity at equilibrium.

Different studies have given slightly different times necessary for reaching long-term equilibrium. Generally, it is five to seven times the longest half-time of the daughters (Cai, 2005; Liu, 2010). From Fig. 1, in the decay series from ^{226}Ra to ^{206}Pb , the

daughter with the longest half-time is ^{210}Pb . Therefore, we can assume that after 110 years (i.e., five times as long as the half-time of ^{210}Pb) to 155 years (i.e., seven times as long as the half-time of ^{210}Pb), ^{226}Ra and its daughter $^{210}\text{Pb}_{\text{eq}}$ reach long-term equilibrium in sediments. In this regard, it is appropriate to take 155 years as the upper limit of ^{210}Pb dating. Otherwise, radioactive decay of $^{210}\text{Pb}_{\text{eq}}$ will follow the law of its precursor ^{226}Ra , whose half-time is 1 602 years. As a consequence, neither Eq. (2) nor Eq. (4) will be appropriate for dating with ^{210}Pb .

More importantly, the upper limit of ^{210}Pb dating is related to the lower limit of detection (*LLD*, unit: Bq) of certain instruments under consideration. Few instruments can distinguish between $^{210}\text{Pb}_{\text{eq}}$ and the residual activity of $^{210}\text{Pb}_{\text{ex}}$ if the latter is less than the *LLD*. According to CNS (1996), the *LLD* of an α spectrometer can be estimated with the following equation:

$$LLD \approx 2KS_0, \quad (9)$$

where S_0 is the standard deviation of measured activities of samples and K is a statistics parameter depending on the confidence level and tolerance (Table 1). Four cases are illustrated in Fig. 2, and it is clear that after four or five half-lives, the residual activity falls within or even below the *LLD* in each case. In such situations, the ^{210}Pb method is unsuccessful at direct dating.

3.2 Reliability of the “prolonging” application for ^{210}Pb dating

As discussed in Section 3.1, a time scale of 100–300 years be-

Table 1. Relationships among K , confidence level and tolerance (CNS, 1996)

α (tolerance)/%	$1-\beta$ (confidence)/%	K
1	99	2.327
2	98	2.054
5	95	1.645
10	90	1.282
20	80	0.842
50	50	0

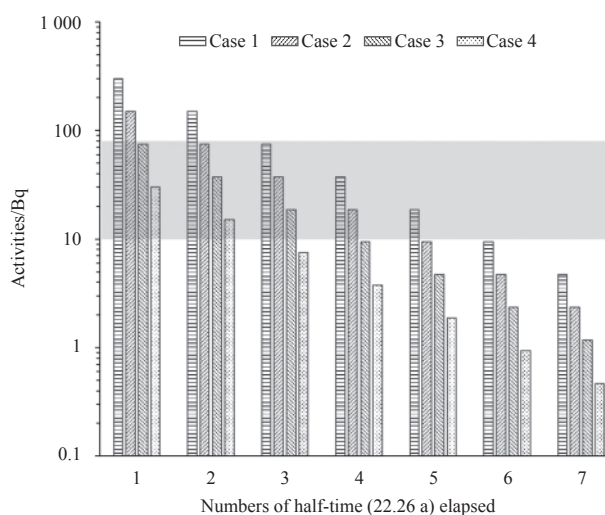


Fig. 2. Illustration of four cases showing the residual activities vs. the lower limit of detection (*LLD*, $\alpha=\beta=0.05$) of an α spectrometer. The grey bar is the *LLD* with the standard deviation of detected activities between 0.05 dpm/g and 0.40 dpm/g. Cases 1, 2, 3 and 4 represent $C_0=10.0$ dpm/g, 5.0 dpm/g, 2.5 dpm/g and 1.0 dpm/g, respectively.

fore the present is beyond the limit of ^{210}Pb dating. It also falls within the error range of ^{14}C dating if 1950 AD is taken as the reference of the “present” or if the Marine Radiocarbon Reservoir Effect is considered (BETA, 2017). Therefore, this time scale is difficult for dating with sediments because no suitable techniques are available. Alternatively, people often calculate century-scale sedimentation rates based on the ^{210}Pb dating technique and then backtrack and give time scales of 200 or 300 years. To determine whether the “prolonging” application is appropriate, we need to answer two questions. How much time is appropriate for the application of ^{210}Pb dating prolonging? How do we know if the prolonged age is correct?

Answers to the first question depend on the characteristics of the sedimentary environments under study. In essence, the prolonging application is an application of “the present is the key to the past”, i.e., assuming that the sedimentary environment in the past several hundred years was generally stable and that the sedimentation rate was generally constant. Only when these conditions are satisfied we may use the latest century-scale sedimentation rate as a scale for measuring the sedimentary history retrospectively. Independent evidence, such as historical records (Zhou et al., 2017), and biomarker or stratigraphic marker methodology (Hall et al., 1999; Donnelly et al., 2001; Eilers et al., 2004; Sawai, 2004) may be helpful in answering the second question. For example, Zhou et al. (2017) reconstructed a 350-year chronicle of typhoon activity at the Hainan Island based on recognition of storm depositions and a retrospective time scale derived from the sedimentation rate, which is estimated using ^{210}Pb dating techniques and checked with historical records (Fig. 3).

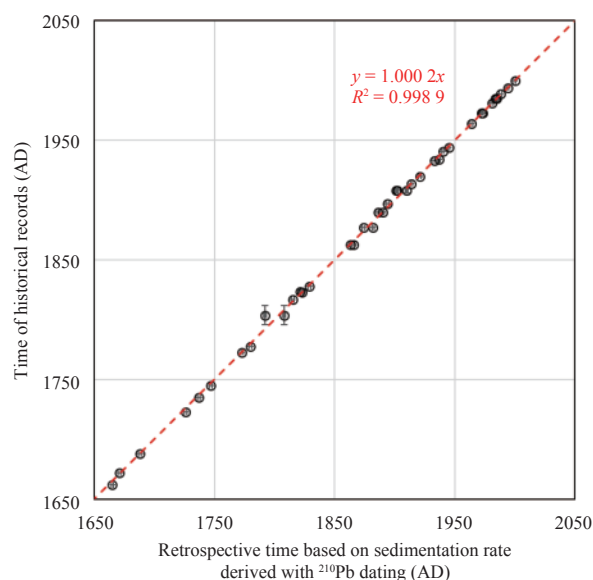


Fig. 3. Retrospective time based on the sedimentation rate derived with ^{210}Pb dating vs. the time recorded in historical literature (data from Zhou et al., 2017).

4 The “loss of age” phenomenon in ^{210}Pb dating

According to incomplete statistics, there are data from nearly 400 cores published since 1980 regarding the modern sedimentation rate on the coast of the Yellow Sea, East China Sea and the adjacent continental shelf (Li et al., 2012). Most of the data were based on ^{210}Pb dating and expressed primarily as the apparent sedimentation rate (i.e., a unit of cm/a was used). We find that when dividing the length of the decay segment of the $^{210}\text{Pb}_{\text{ex}}$ pro-

file of the cores (i.e., above background) by the apparent sedimentation rates, the result is often less than 100 years (Fig. 4) with a mean value of only 40 ± 20 years (Fig. 5). This finding indicates that when the ^{210}Pb dating techniques that can nominally date the century-scale are applied to modern marine sedimentary environments, they record a sedimentary history of less than 100 years. Through analysis, we suggest the following four possible reasons.

4.1 Sediment compaction effect

If we directly divide the length of a decay segment of a core by the apparent sedimentation rate to calculate the age period, the implied assumption of this data processing is that the bulk density of the sediment from the top to the bottom of the core is constant. In fact, due to the compaction effect (including natural compaction and mechanical compaction during sampling), the porosity of sediments in cores gradually decreases from the sur-

face to the bottom, and the corresponding bulk density gradually increases. From the published literature, the bulk density of the surface sediments in marine environments is primarily between 0.5 and 0.9 g/cm^3 (Flemming et al., 2000), whereas the bulk density at the bottom layer of cores is primarily between 1.1 and 1.3 g/cm^3 . Therefore, using a single bulk density value for depth correction significantly increases the corrected length of cores (Zou et al., 1982). Figure 6 shows three cores collected from the East China Sea. After correction, the length of the cores increases by $1/4$ to $1/2$.

In fact, the sediment flux ($\text{g}/(\text{cm}^2 \cdot \text{a})$) is the best way to represent the sedimentation rate. It can effectively avoid the calculation error caused by the compaction effect and the variation in sediment bulk density. Fan et al. (2000) used published data and gave the following empirical equations to derive the mass depth of a complete core from the water content of surface sediment under the condition of continuous deposition:

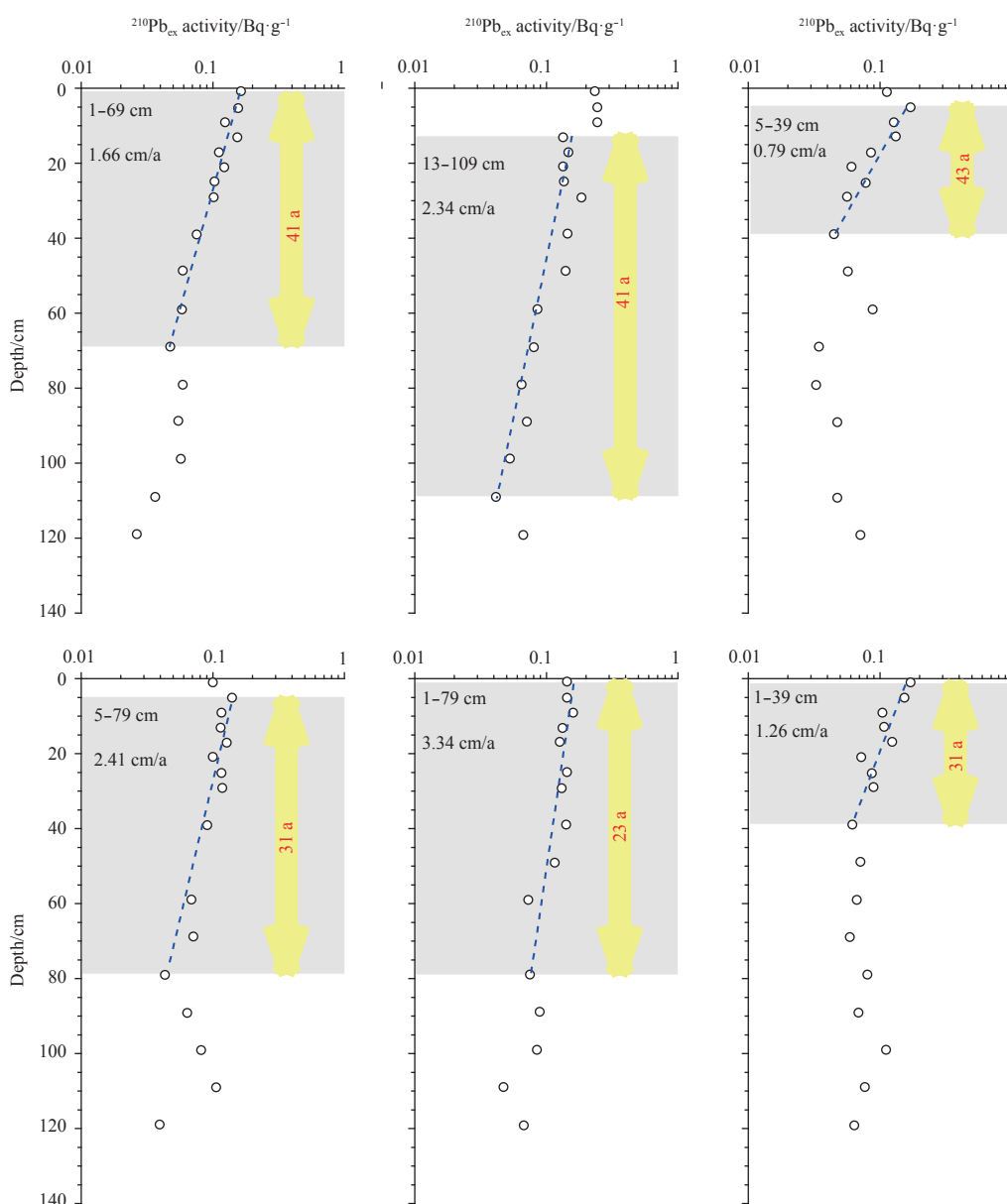


Fig. 4. One literature case showing that the periods derived with the apparent sedimentation rate are much less than 100 years (data from Liu et al., 2009).

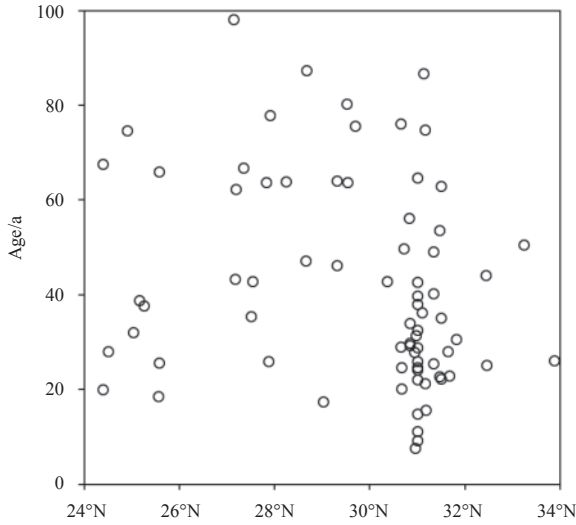


Fig. 5. Data of short cores sampled in the inner shelf of the East China Sea. The age of each core is derived by dividing the length (cm) of the decay segment of the $^{210}\text{Pb}_{\text{ex}}$ profile by its apparent sedimentation rate (cm/a).

$$P = p_1 e^{-k_1 x}, \quad (10)$$

$$Z = \int_0^x \rho_s (1 - p_1 e^{-k_1 x}) dx = \rho_s \left[x - \frac{p_1}{k_1} (1 - e^{-k_1 x}) \right], \quad (11)$$

where P is sediment porosity, Z is mass depth (g/cm^2), x is natural depth (cm), ρ_s is sediment bulk density, p_1 is surface sediment porosity, and k_1 is an empirical constant related to regional sedimentary environment characteristics. k_1 can be obtained by fitting Eq. (9) to the depth variation of the measured porosity of sediment in cores. For example, for the mud zone in the East China Sea, k_2 is 0.005 (Fan et al., 2000). Thus, through Eq. (10), we can conveniently achieve compaction correction.

4.2 Effect of instrument measurement accuracy on the judgment of the background layer

In actual studies, researchers have primarily used Procedure II to determine the ^{210}Pb background. From published studies, the background $^{210}\text{Pb}_{\text{eq}}$ in inner shelf muds of the Yellow Sea and East China Sea is approximately 1.0 dpm/g, whereas the $^{210}\text{Pb}_{\text{ex}}$ of surface sediments is 1–10 dpm/g. Figure 7 illustrates an example under ideal conditions. With ages of sediments increasing downward, the $^{210}\text{Pb}_{\text{total}}$ specific activity value decreases due to $^{210}\text{Pb}_{\text{ex}}$ decay. Comparing the four curves in Fig. 7, we find that the smaller the surface $^{210}\text{Pb}_{\text{ex}}$ is, the faster it approaches $^{210}\text{Pb}_{\text{eq}}$, and the difference between $^{210}\text{Pb}_{\text{eq}}$ and $^{210}\text{Pb}_{\text{ex}}$ also becomes more difficult to distinguish. Figure 8 is another illustration similar to Fig. 7. We randomized the errors of the $^{210}\text{Pb}_{\text{total}}$ measurements of different ages (–5% to +5%). As shown, for cores with a small specific activity, after two to three half-lives, analytical instruments can scarcely distinguish the difference between the $^{210}\text{Pb}_{\text{eq}}$ and the $^{210}\text{Pb}_{\text{total}}$. The estimated age of the $^{210}\text{Pb}_{\text{eq}}$ layer may decrease by a large amount. For example, when the surface $^{210}\text{Pb}_{\text{ex}}$ is 1.0 dpm/g and 2.5 dpm/g, the corresponding age of the assigned $^{210}\text{Pb}_{\text{eq}}$ is only 60 years and 80 years, respectively (Fig. 8).

In fact, the examples shown in Fig. 7 are ideal cases. Limited by the analytical capabilities of laboratory instruments and by research funds, many early ^{210}Pb dating data were unevenly measured and scarcely distributed along the depths of cores. If the measurement interval for ^{210}Pb activity at the lower parts of cores is greater than 10 cm, the error in judging the background layer is larger. Therefore, we suggest that under the present circumstances in which laboratory analytical capabilities and research funds have increased significantly, the measurement interval of ^{210}Pb activity should be reduced as much as possible, and it is best to use uniform intervals.

4.3 Selection of the ^{210}Pb background value and its effect on dating results

We find that a small difference in the ^{210}Pb background can cause a significant discrepancy in the calculated sedimentation

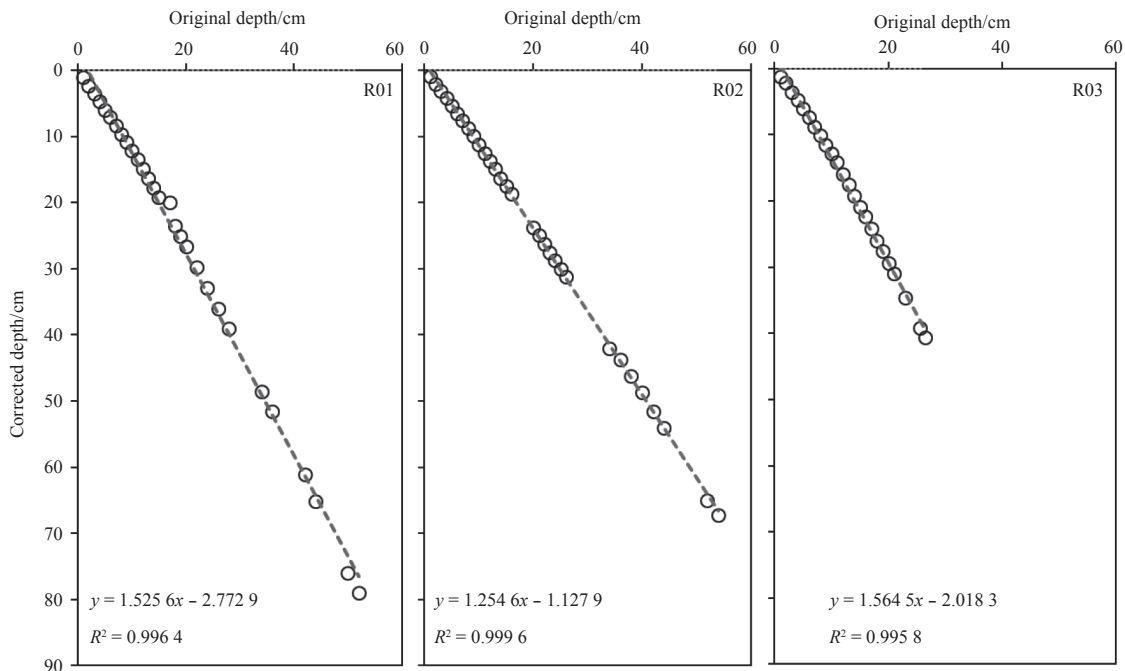


Fig. 6. Depth correction for three cores sampled in the East China Sea (data from Zou et al., 1982).

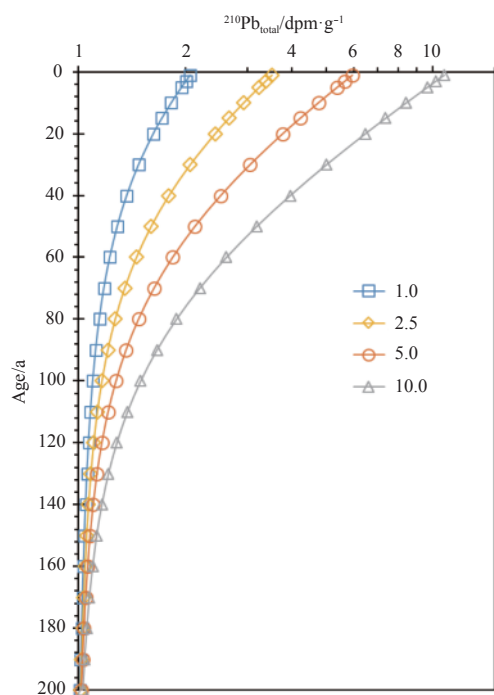


Fig. 7. Decay of the surface activity ($^{210}\text{Pb}_{\text{total}}$) with time in different cores. $^{210}\text{Pb}_{\text{eq}}$ is 1.1 dpm/g, and four curves represent the age when the surface $^{210}\text{Pb}_{\text{ex}}$ is 1.0, 2.5, 5.0 and 10.0 dpm/g.

rate and the recorded age limit of the ^{210}Pb decay segment. Figure 9 shows a core taken in the East China Sea, with a typical two-segment vertical distribution of ^{210}Pb specific activity. The top slope segment reflects the decay process of $^{210}\text{Pb}_{\text{ex}}$. The bottom straight segment reflects the condition of $^{210}\text{Pb}_{\text{eq}}$. The back-

grounds obtained by two procedures mentioned in Section 3.1 are 1.1 dpm/g and 1.2 dpm/g, respectively. Although their difference is less than 10%, the calculated age periods of the slope segment using three combinations of backgrounds and dating models differ by 20 years. CRS model with larger value of background activities will give elder dating results than with smaller background activities, and dating results derived with CRS model are generally older than with CIC model.

To reduce the effect of the ^{210}Pb background value on the dating result as much as possible, we suggest measuring the ^{226}Ra and ^{210}Pb activity at the same time, which can effectively avoid the uncertainty error brought by the above empirical selection.

4.4 Disturbance, erosion and migration effects

Taking the Zhe-Min inner shelf mud zones as examples, the sediments there primarily originate from a portion of Changjiang River sediments transferred to the sea that are transported southward by the Zhe-Min coastal current (Qin et al., 1996; Liu et al., 2006; Gao and Collins, 2014). Summer typhoons, winter waves and strong winds occur frequently, which can disturb sediments and produce erosional transportation (Dai, 1992; Xie et al., 2001). In other words, the behavior of ^{210}Pb in the Zhe-Min inner shelf mud zones can hardly satisfy the condition required by the ^{210}Pb dating model (Appleby and Oldfield, 1983; Wan, 1997). Once there is an extreme weather event, sedimentary records are lost and reflect a “loss of age” of ^{210}Pb records in cores.

5 Discussion

5.1 Suitability of the ^{210}Pb dating techniques in marine environments

With the CIC model or the CRS model, the use of the ^{210}Pb dating techniques must follow several basic assumptions (Wan, 1997). (1) Sediments must be a closed system, and their source,

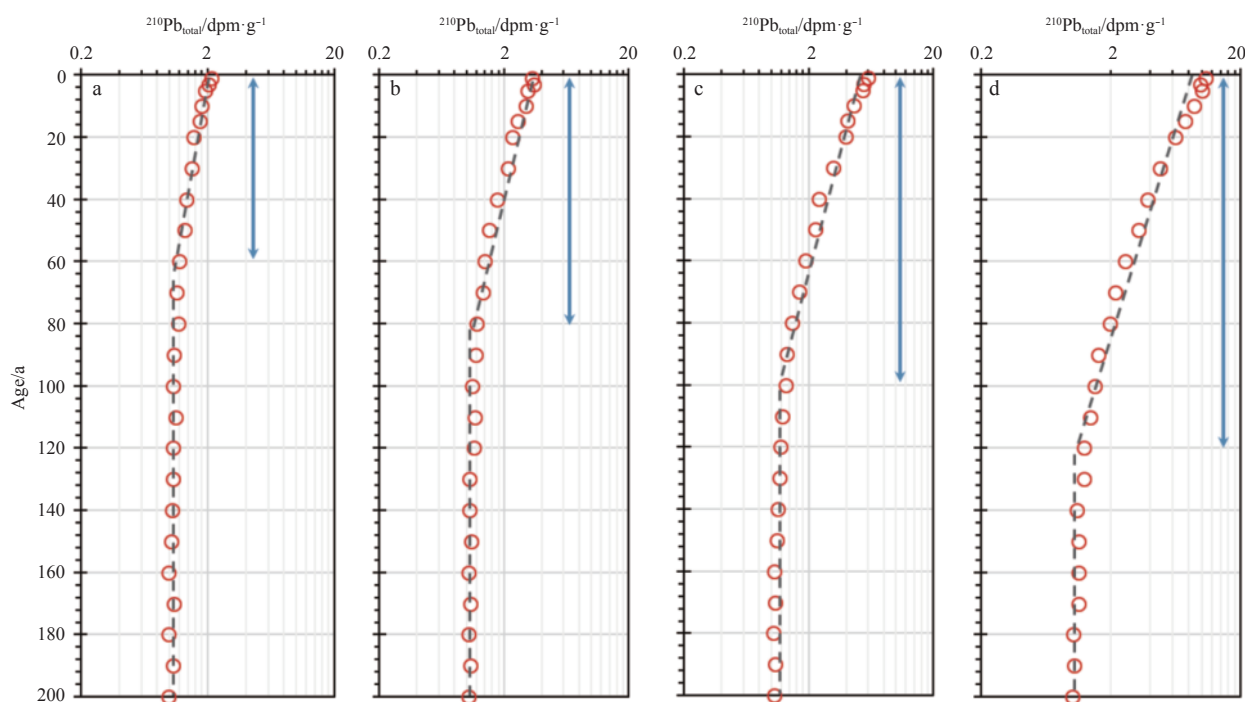


Fig. 8. Determining background layers with the same $^{210}\text{Pb}_{\text{eq}}$ but different surface $^{210}\text{Pb}_{\text{ex}}$ values. Assuming a stable sedimentary environment is studied, the background ^{226}Ra activity is 1.1 dpm/g, and the surface $^{210}\text{Pb}_{\text{ex}}$ values are 1.0 dpm/g (a), 2.5 dpm/g (b), 5.0 dpm/g (c) and 10.0 dpm/g (d).

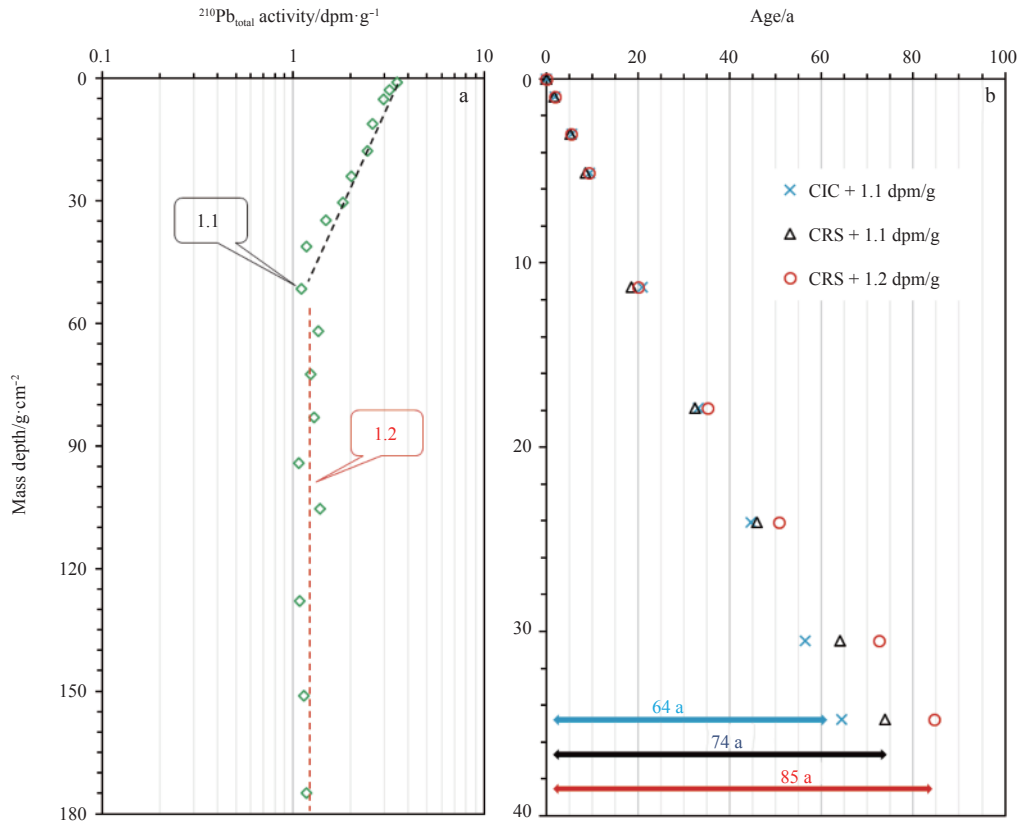


Fig. 9. The ^{210}Pb specific activities profile of a short core collected in the East China Sea (a) and dating results of three combinations of background values and dating models (b). Left panel: the red label indicates the value of $^{210}\text{Pb}_{\text{eq}}$ determined with Procedure I, whereas the black label indicates the value of $^{210}\text{Pb}_{\text{eq}}$ determined with Procedure II (see the text in Section 3.1 for explanation). Right panel: blue cross, black triangle and red cycle represent combinations of CIC model ($^{210}\text{Pb}_{\text{eq}}=1.1$ dpm/g), CRS model ($^{210}\text{Pb}_{\text{eq}}=1.1$ dpm/g) and CRS model ($^{210}\text{Pb}_{\text{eq}}=1.2$ dpm/g), respectively. The corresponding dating results for the layer of background are 64 years, 74 years and 85 years.

accumulation rate and ^{210}Pb input flux must be stable. (2) Compared to the residence time in water, ^{210}Pb that enters a lake or bay is effectively transferred to sediments. (3) The $^{210}\text{Pb}_{\text{eq}}$ in sediments should be in equilibrium with its precursor ^{226}Ra . (4) The ^{210}Pb accumulated in sediments does not migrate after deposition. Experience with ^{210}Pb dating in different geographic settings indicates that with the increasing openness of sedimentary environments and increasing possibility of post-sedimentation disturbances by environmental factors, the above four conditions are more difficult to satisfy.

In a shallow continental shelf environment, rivers that go into the sea bring a large amount of suspended sediments (at least in the delta region of large rivers) and may become a non-negligible ^{210}Pb source besides atmospheric deposition (Gao et al., 2017). Furthermore, surface sediments in marine environments are often mobile and easily transported under the influence of tides, waves and biological disturbances. Thus, it is difficult to satisfy the two conditions for applying the CRS model: “material source, accumulation rate and ^{210}Pb input flux are all stable” and “the ^{210}Pb accumulated in sediments does not migrate after deposition”. Therefore, we believe that in an open and shallow environment of a continental shelf, the suitability conditions of the ^{210}Pb dating techniques cannot be strictly satisfied. As Appleby and Oldfield (1983) noted, we first need to carefully analyze the sedimentary environmental characteristics of the study region and select an appropriate dating model. At the same time, we

need to use other independent dating methods and sedimentology methods to verify the result of ^{210}Pb dating.

5.2 Value of A_0 in marine sedimentary dynamics

We discussed earlier that the CRS model requires that the total accumulated $^{210}\text{Pb}_{\text{ex}}$ in the same or equivalent sedimentary environments should reflect the local atmospheric $^{210}\text{Pb}_{\text{ex}}$ deposition flux (Appleby and Oldfield, 1983). According to this assumption, we can infer that the total accumulative $^{210}\text{Pb}_{\text{ex}}$ in sediments should be 12.66–66.11 dpm/cm² (i.e., the product of the atmospheric deposition flux and the average lifetime of ^{210}Pb ; 1 Bq=60 dpm), knowing that the global atmospheric $^{210}\text{Pb}_{\text{ex}}$ deposition flux is 0.18–0.94 Bq/(m²·d) (Liu, 2010). Oldfield et al. (1978) found substantial evidence from two cores taken in a lake in that although their sedimentation rate differed by three times, the total accumulative $^{210}\text{Pb}_{\text{ex}}$ was essentially the same.

From the analysis in Sections 5.1 and 5.2, it is difficult for marine sediments to satisfy the requirements of the CRS model. However, this situation may bring new information to marine sedimentary studies. Using the core shown in Fig. 9 as an example, the two-segment ^{210}Pb profile lacks an upper mixing layer. In the past, this phenomenon was often interpreted as a weak mixing and weak disturbance in the water-sediment interface. However, if the core once had a mixing layer but was eroded, it would also form a two-segment ^{210}Pb profile. How do we determine which explanation is closer to reality? We analyzed the rela-

tion between A_0 (the total accumulative $^{210}\text{Pb}_{\text{ex}}$ in sediments) and the atmospheric $^{210}\text{Pb}_{\text{ex}}$ deposition flux in the same period; we found that they are essentially equal. Thus, we could infer that the sedimentary environment of this core was stable and lacked any post-deposition disturbance, resuspension and erosion.

6 Conclusions

(1) Direct ^{210}Pb dating works best for a time scale with an upper limit of 110–155 years. The “prolonging” application is acceptable only if the sedimentary environment in the past several hundred years was stable and the sedimentation rate was generally constant, and verification with independent evidence (such as historical records or biomarker methodology) is needed.

(2) Due to the compaction effect that occurs during deposition and sampling, the widely used “apparent sedimentation rate” (cm/a) will cause inherent and systematic errors in ^{210}Pb dating. We recommend the sediment mass flux ($\text{g}/(\text{cm}^2\cdot\text{a})$) instead of the apparent sedimentation rate (cm/a) for ^{210}Pb dating.

(3) In addition to the compaction effect, the uncertainty of activity measurement and the post-deposition erosion and migration can also lead to “loss of age” in ^{210}Pb dating. Reducing the interval of sub-samples for ^{210}Pb activity measurement and measuring the ^{226}Ra activity concurrently can effectively lessen the ^{210}Pb dating error.

(4) Open marine environments cannot strictly satisfy the requirements for the ^{210}Pb dating model. However, from the aspect of marine environment characteristics, ^{210}Pb dating data may provide useful information on issues such as the material sources of marine sediments and the post-deposition erosion and disturbance.

Acknowledgements

The authors are grateful to Du Jinzhou and Liu Zhiyong and Zhou Liang for their insightful comments and helpful information.

References

- Alexander C R, Demaster D J, Nittrouer C A. 1991. Sediment accumulation in a modern epicontinental-shelf setting: the Yellow Sea. *Marine Geology*, 98(1): 51–72
- Appleby P G, Oldfield F, Thompson R, et al. 1979. ^{210}Pb dating of annually laminated lake sediments from Finland. *Nature*, 280(5717): 53–55
- Appleby P G, Oldfield F. 1978. The calculation of lead-210 dates assuming a constant rate of supply of unsupported ^{210}Pb to the sediment. *Catena*, 5(1): 1–8
- Appleby P G, Oldfield F. 1983. The assessment of ^{210}Pb data from sites with varying sediment accumulation rates. *Hydrobiologia*, 103(1): 29–35
- BETA (Beta Analytic Radiocarbon Dating). 2017. Calibration of Carbon 14 Dating Results. <https://www.radiocarbon.com/calendar-calibration-carbon-dating.htm> [2014-05-15/2017-12-20]
- Binford M W. 1990. Calculation and uncertainty analysis of ^{210}Pb dates for PIRLA project lake sediment cores. *Journal of Paleolimnology*, 3(3): 253–267
- Cai Chonggui. 2005. A proof of the secular equilibrium formula in radioactive cascade decay series. *College Physics* (in Chinese), 24(8): 44–46
- Chen Fei, Yin Liangliang, Kong Xiangyin, et al. 2016. Uncertainty evaluation of strontium-90 analysis in biological samples. *Journal of Radiation Research and Radiation Processing* (in Chinese), 34(1): 59–64
- Chen Zhongyuan, Saito Y, Kanai Y, et al. 2004. Low concentration of heavy metals in the Yangtze estuarine sediments, China: a diluting setting. *Estuarine, Coastal and Shelf Science*, 60(1): 91–100
- CNS (China National Standard). 1996. Analytical method for radionuclides by alpha spectrometry (GB/T 16141–1995). <http://www.nhfpc.gov.cn/ewebeditor/uploadfile/2014/10/20141030160815598.pdf> [2014-10-30/2017-12-20]
- Dai Zeheng. 1992. Comprehensive Survey Report on the Coastal Zone and Coastal Resources in Zhejiang Province (in Chinese). Beijing: China Ocean Press, 121–130
- Demaster D J, Mckee B A, Nittrouer C A, et al. 1985. Rates of sediment accumulation and particle reworking based on radiochemical measurements from continental shelf deposits in the East China Sea. *Continental Shelf Research*, 4(1–2): 143–158
- Donnelly J P, Bryant S S, Butler J, et al. 2001. A 700 yr sedimentary record of intense hurricane landfalls in southern New England. *GSA Bulletin*, 113(6): 714–727
- Eilers J M, Kann J, Cornett J, et al. 2004. Paleolimnological evidence of change in a shallow, hypereutrophic lake: Upper Klamath Lake, Oregon, USA. *Hydrobiologia*, 520(1–3): 7–18
- Fan Dejiang, Yang Zuosheng, Guo Zhigang. 2000. Review of ^{210}Pb dating in the continental shelf of China. *Advance in Earth Sciences* (in Chinese), 15(3): 297–302
- Flemming B W, Delafontaine M T. 2000. Mass physical properties of muddy intertidal sediments: some applications, misapplications and non-applications. *Continental Shelf Research*, 20(10–11): 1179–1197
- Gao Jianhua, Jia Jianjun, Sheng Hui, et al. 2017. Variations in the transport, distribution, and budget of ^{210}Pb in sediment over the estuarine and inner shelf areas of the East China Sea due to Changjiang catchment changes. *Journal of Geophysical Research: Earth Surface*, 122(1): 235–247
- Gao S, Collins M B. 2014. Holocene sedimentary systems on continental shelves. *Marine Geology*, 352: 268–294
- Goldberg E D. 1963. Geochronology with ^{210}Pb . In: *Radioactive Dating, Proceedings of the Symposium on Radioactive Dating Held by the International Atomic Energy Agency in Co-operation with the Joint Commission on Applied Radioactivity*. Athens: International Atomic Energy Agency, 121–131
- Hall R I, Leavitt P R, Quinlan R, et al. 1999. Effects of agriculture, urbanization, and climate on water quality in the northern Great Plains. *Limnology and Oceanography*, 44(3 Part 2): 739–756
- Jia Jianjun, Gao Jianhua, Liu Yifei, et al. 2012. Environmental changes in Shamei Lagoon, Hainan Island, China: Interactions between natural processes and human activities. *Journal of Asian Earth Sciences*, 52: 158–168
- Jin Xianglong. 1992. *Submarine Geology of East China Sea*. Beijing: China Ocean Press (in Chinese), 524
- Koide M, Soutar A, Goldberg E D. 1972. Marine geochronology with ^{210}Pb . *Earth and Planetary Science Letters*, 14: 442–446
- Li Fengye, Shi Yulan, Shen Shunxi, et al. 1996. Isotopic record of modern sedimentary environment in the South Yellow Sea. *Oceanologia et Limnologia Sinica* (in Chinese), 27(6): 584–589
- Li Jun, Hu Bangqi, Dou Yanguang, et al. 2012. Modern sedimentation rate, budget and supply of the muddy deposits in the East China Seas. *Geological Review* (in Chinese), 58(4): 745–756
- Liu Guangshan. 2010. *Isotopic Oceanography* (in Chinese). Zhengzhou: Zhengzhou University Press, 1–9
- Liu Shengfa, Shi Xuefa, Liu Yanguang, et al. 2009. Sedimentation rate of mud area in the East China Sea inner continental shelf. *Marine Geology & Quaternary Geology* (in Chinese), 29(6): 1–6
- Nittrouer C A, Sternberg R W, Carpenter R, et al. 1979. The use of ^{210}Pb geochronology as a sedimentological tool: application to the Washington continental shelf. *Marine Geology*, 31: 296–316
- Oldfield F, Appleby P G, Battarbee R W. 1978. Alternative ^{210}Pb dating: results from the New Guinea Highlands and Lough Erne. *Nature*, 271(5643): 339–442
- Oldfield F, Appleby P G. 1984. Empirical testing of ^{210}Pb dating models for lake sediments. In: Haworth E T, Lund J W G, eds. *Lake Sediments and Environmental History*. Leicester: Leicester University Press
- Qian Jiangchu, DeMaster D J, Nittrouer C A, et al. 1985. Geochemistry features of Pb-210 in the estuary of the Changjiang River and its adjacent shelf. *Acta Sedimentologica Sinica* (in Chinese),

- 3(4): 31–44
- Qin Yunsan, Zhao Yiyang, Chen Lirong, et al. 1996. Geology of the East China Sea. Beijing: Science Press
- Robbins J A. 1978. Geochemical and geophysical applications of radioactive lead. In: Nriagu J O, ed. Biogeochemistry of Lead in the Environment. Amsterdam: Elsevier Scientific, 285–393
- Sanchez-Cabeza J A, Ruiz-Fernández A C. 2012. ^{210}Pb sediment radiochronology: An integrated formulation and classification of dating models. *Geochimica et Cosmochimica Acta*, 82: 183–200
- Sawai Y. 2002. Evidence for 17th-century tsunamis generated on the Kuril–Kamchatka subduction zone, Lake Tokotan, Hokkaido, Japan. *Journal of Asian Earth Sciences*, 20(8): 903–911
- Su Xianze, Ma Wentong, Xu Shengli, et al. 1984. The method of lead-210 geochronology for marine sediments. *Taiwan Strait (in Chinese)*, 3(1): 50–58
- Wan Guojiang. 1997. ^{210}Pb Dating for recent sedimentation. *Quaternary Sciences (in Chinese)*, 17(3): 230–239
- Xia Xiaoming, Xie Qinchun, Li Yan, et al. 1999. ^{137}Cs and ^{210}Pb profiles of the seabed cores along the East China Sea coast and their implications to sedimentary environment. *Donghai Marine Science (in Chinese)*, 17(4): 20–27
- Xia Xiaoming, Yang Hui, Li Yan, et al. 2004. Modern sedimentation rates in the contiguous sea area of Changjiang Estuary and Hangzhou Bay. *Acta Sedimentologica Sinica (in Chinese)*, 22(1): 130–135
- Xie Qinchun, Ma Liming, Li Bogen, et al. 2001. Rapid storm sedimentation of Maotou deep trough in the Sanmen Bay of Zhejiang. *Haiyang Xuebao (in Chinese)*, 23(5): 78–86
- Zhang Jing, Mou Dehai, Du Jinzhou, et al. 2008. Study on comparison of excess ^{210}Pb chronology of several models. *Marine Environmental Science (in Chinese)*, 27(4): 370–374, 382
- Zhang Rui, Pan Shaoming, Wang Yaping, et al. 2009. Sedimentation rates and characteristics of radionuclide ^{210}Pb at the subaqueous delta in Changjiang Estuary. *Acta Sedimentologica Sinica (in Chinese)*, 27(4): 704–713
- Zhou Liang, Gao Shu, Yang Yang, et al. 2017. Typhoon events recorded in coastal lagoon deposits, southeastern Hainan Island. *Acta Oceanologica Sinica*, 36(4): 37–45
- Zou Hanyang, Su Xianze, Yu Xingguang, et al. 1982. Determination of recent sedimentation rates on the continental shelf of the East China Sea using ^{210}Pb method. *Journal of Oceanography in Taiwan Strait (in Chinese)*, 1(2): 30–40