



Clay Mineralogical Records in the North Bohai Coast of China in the Last Century: Sediment Provenance and Morphological Implications

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Provenance identification of the clastic sediments is critical for paleoenvironmental and morphological reconstruction on the estuarine coast. The North Bohai Coast (NBC) receives tremendous sediments mainly from the Liaohe and Daling fluvial systems. However, the detailed morphological evolution is still unclear due to the lack of precise discrimination of the sediment provenance. In this study, the clay mineralogy of the coastal sediments in the NBC was investigated to determine variation of the fine-grained sediment sources in the last century. The clay mineral content in the modern riverine sediments in the NBC was used to model sedimentary contribution from different fluvial systems. ²¹⁰Pb and ¹³⁷Cs radioactivity was measured to constrain the timing of centennial marine–terrestrial sedimentation. The migration of coastline and river channels in the NBC during the last hundred years was reconstructed by digitalizing the historical charts and satellite image. ²¹⁰Pb following the CIC model and ¹³⁷Cs dating are considered not applicable for the dated cores, whilst ²¹⁰Pb using the CRS model yields acceptable chronologic results. Non-linear modeling results of the clay mineral composition are consistent with the lithological and coastal geomorphologic constraints. The outcome of provenance discrimination indicates that clay minerals in western and eastern NBC are mainly derived from the Daling and Liaohe rivers, respectively. In the middle NBC, the Daling River input was relatively major during the 1910s, 1940s, 1980s, and 2000s. The increase of Daling River input in the first two periods resulted from progradation and river channel migration, respectively. The fluvial contribution from the Liaohe River was major in the middle NBC during 1920–1940 and 1950–1970 AD, which attributes to rapid progradation of the Liaohe River and channel migration of the Daling River, respectively.

Keywords: clay mineralogy, provenance, historical charts, non-linear programming modeling, paleoenvironmental change, North Bohai Coast

INTRODUCTION

The estuarine coast is a critical area due to its ecological significance, environmental vulnerability, and socio-economic importance (IPCC, 2014). Paleoenvironment and morphology in the estuarine coast have changed significantly in different timescales due to the natural and anthropogenic impacts. It is necessary to reconstruct the history of coastal environmental evolution to improve our understanding of regional response in the estuary to global change and for better protection of the estuarine coast.

River channel migration and sediment flux alternation are the two crucial controlling factors in shaping the estuarine morphology, which could be revealed by the provenance variation of the estuarine coastal sediments, especially for those controlled by multiple rivers. Physical, chemical, and mineralogical characteristics of the sediments have been used to trace sediment sources (e.g., Biscay, 1965; Jacobsen and Wasserburg, 1980; Chamley, 1989; Liu et al., 2003; Colin et al., 2006; Kissel et al., 2016; Gray et al., 2019). Among them, clay mineralogy is thought to be applicable in tracing the sediment provenance for the marine sediments, which has been widely used in the east margin of the Asian Continent (e.g., Hu et al., 2014; Cho et al., 2015; Liu et al., 2018; Cai et al., 2020). The previous studies have mainly focused on the provenance of multiple sediment sources in the marginal seas. Nevertheless, the estuarine coast has been rarely investigated, which largely limits us in reconstructing the history of coastal geomorphology evolution precisely and/or understanding the natural and anthropogenic control on the estuarine coasts over different timescales.

The northern coast of the Bohai Sea is one of the largest coastal estuaries in eastern China, with considerable sediments deriving from the Liaohe and Daling rivers (IOCAS, 1985). Owing to the paleoenvironmental and ecological significance, the sediments in the North Bohai Coast (NBC) and the adjacent area have been widely explored (e.g., Chen et al., 2010; Dou et al., 2014; He et al., 2018; Li J. et al., 2018; Li H. et al., 2019; Liu et al., 2019; Liu et al., 2020; Wang et al., 2020). Coastal geomorphology has significantly changed with frequent river channel migration and decreasing fluvial sediment flux in the NBC over the last century (Chen et al., 2010; Li Y. et al., 2018). Consequently, as one of the largest estuarine wetlands in eastern China, the NBC is facing the threats of wetland degradation and ecological destruction (Zhou et al., 2016). However, the influence of the Daling and Liaohe fluvial systems on the morphological change in the aspect of sedimentation supply is still unclear. The detailed morphological evolution caused by river channel migration has not been well investigated either. Since the sediments have recorded the information of provenance, it is meaningful to reconstruct the coastal morphology by discriminating the sediment provenance precisely in the NBC over the last century. It is also significant for a better understanding of the natural-anthropogenic influences on the coastal sedimentary processes.

The outcome of our previous investigation showed that the clay mineral assemblage in the modern Liaohe and Daling riverine sediments is illite and smectite dominated, respectively (Liu et al., 2019). This implies that clay mineralogy is potentially useful to trace the sources of sediments deposited in the NBC. Moreover, frequent river channel migration and continuous coastline retreats over the last century have been well documented in the historical archives. Therefore, the reliability of the discriminated provenance of the sediments in the NBC over the last century using clay mineralogy could be possibly assessed by the information on fluvial and coastline migration documented in the historical archives.

In this study, the shallow core sediments collected from the NBC were used for clay mineralogy, chronology, and stratigraphy studies. ^{210}Pb and ^{137}Cs activities for the sediments from four cores were measured to quantify the sedimentation rate. X-ray diffraction (XRD) analysis was carried out to determine the composition of the four main clay minerals, i.e. smectite, illite, kaolinite, and chlorite, for the sediments from ten representative cores. The historical maps were digitalized to extract information on the coastline and river channel migration in the last century. The main objectives of this study are as follows. First, we aim to construct a model to quantify the sediment contributions from the Liaohe and Daling rivers based on the clay mineralogy of modern riverine sediments (Liu et al., 2019). The reliability of the model was assessed by comparing the modeling result of the core sediments and the historical river channel migration. Our second aim is to reconstruct sediment transport and morphological evolution in the NBC over the last century combining the clay mineralogy and historical coastline-river channel migration.

STUDY AREA

The NBC is the marine-terrestrial interacted area in the Liaodong Bay, northern Bohai Sea (**Figure 1**). The NBC receives tremendous terrigenous sediments transported mainly by the Liaohe and Daling rivers. The mean tidal range in the NBC is 2.6 m. The annual mean temperature and precipitation in the study area are 8.9°C and 667.4 mm, respectively.

The Liaohe River is one of the largest rivers in China, originating from the southwest end of the Greater Khingan Mountains and the Changbai Mountain (**Figure 1A**). The drainage basin of the Liaohe River covers $2.20 \times 10^5 \text{ km}^2$, including the local rivers, such as the Daliao and Raoyang rivers. The annual runoff of the Liaohe River is *ca.* $3.19 \times 10^9 \text{ m}^3$. The main bedrock type in the upper Liaohe River Basin contains Mesozoic sedimentary rock and granite, Precambrian volcanic, and sedimentary rock. The middle-lower Liaohe River Basin is covered by the thick Quaternary sediments (**Figure 1A**, Liaoning Geology Bureau, 1983; IOCAS, 1985).

The Daling River originates from northern Yanshan Mountain. The area of the drainage basin is $2.32 \times 10^4 \text{ km}^2$. With a sediment flux of 13.53 Mt/a and a mean suspended sediment concentration of 17.0 kg/m^3 , the Daling River is regarded as one of the muddiest rivers worldwide (Milliman and Syvitski, 1992). Bedrock types in the upper-middle Daling River Basin mainly include Mesozoic volcanic and sedimentary rocks, as well as Precambrian and Paleozoic volcanic rocks. The lower Daling River Basin is mainly covered by quaternary sediments (**Figure 1A**).

Due to Early-Middle Holocene sea-level rise, the lower Liaohe—Daling River Basin was submerged under seawater. The created accommodation space was infilled by terrigenous sediments afterward. In this study, the investigated area of the modern NBC was divided into the west, middle and east sub-zones by the Liaohe and Daling river channels (**Figure 1B**).

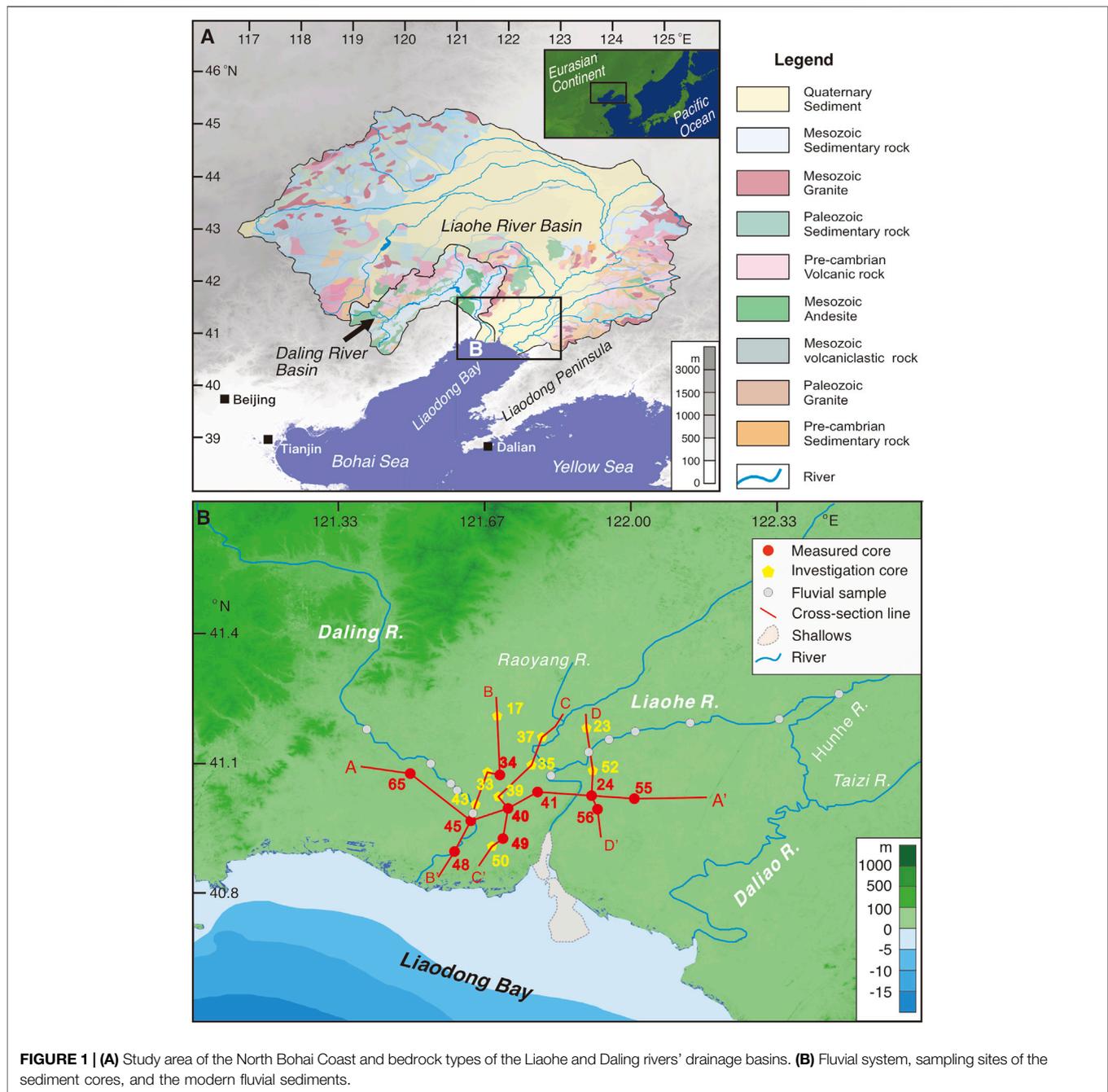


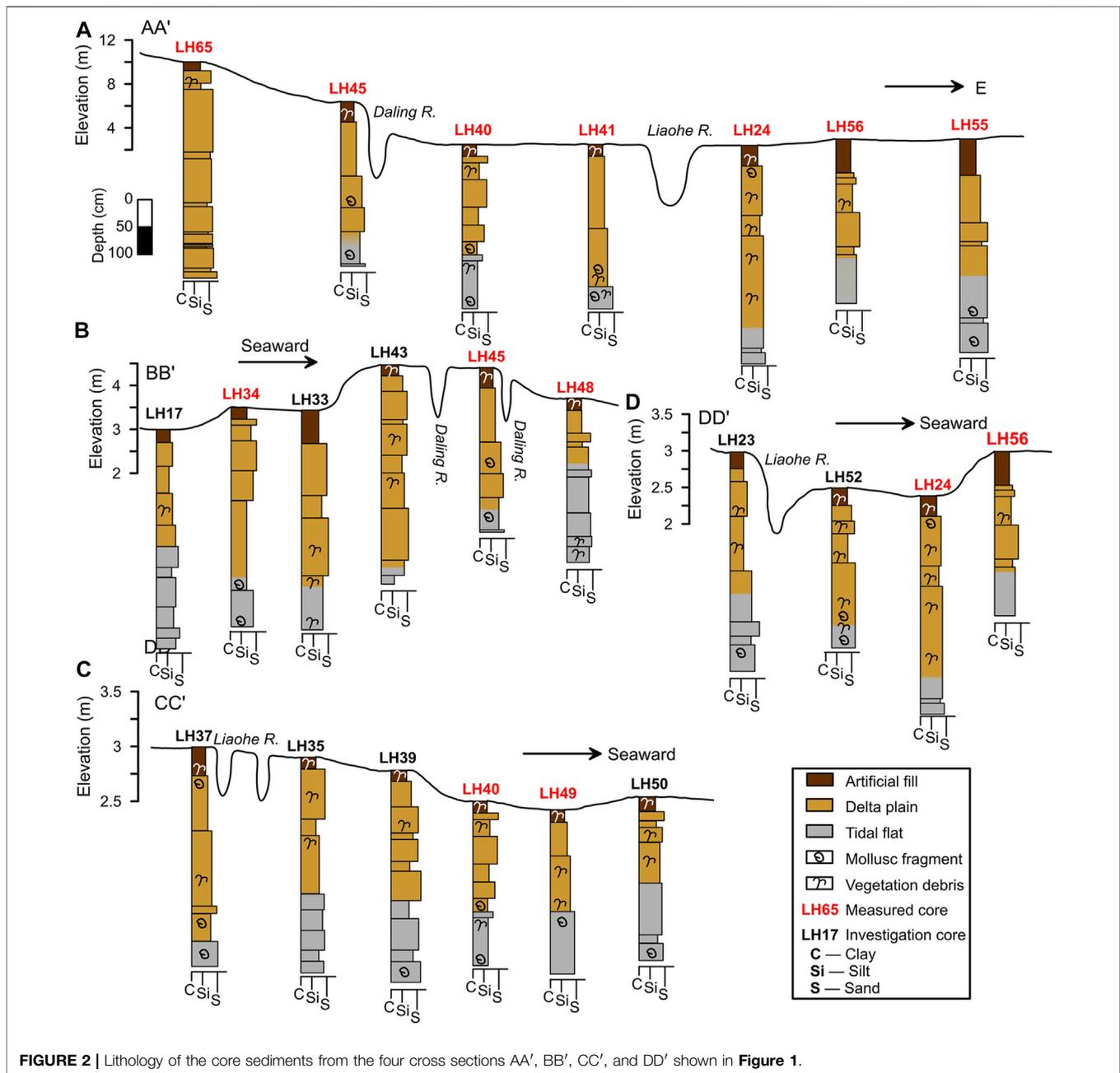
FIGURE 1 | (A) Study area of the North Bohai Coast and bedrock types of the Liaohe and Daling rivers' drainage basins. **(B)** Fluvial system, sampling sites of the sediment cores, and the modern fluvial sediments.

MATERIALS AND METHODS

Sample Collection

Nineteen cores were collected in the modern NBC using the portable power coring system in 2017. The lengths of the cores range from 274 to 400 cm. All the cores were photographed and lithologically described in the field. The cores are shown in five cross sections (AA'—DD') in **Figure 1** and **Figure 2**. The sediment cores can generally be vertically divided into two parts, including the lower dark-gray clayey silt and the upper brownish silty clay. The lower part contains

mollusk shell fragments, whilst vegetation fragments were generally found in the upper part. A transitional layer of brownish-gray clayey silt with a length of 10–30 cm generally exists, which indicates that the environment gradually changed from intertidal flat to delta plain in the NBC. In total, 702 samples from ten cores were sampled and measured for clay mineralogical X-ray diffraction (XRD) analysis with an interval of 4 cm (red filled circles in **Figure 1B**). ^{210}Pb and ^{137}Cs dating were conducted using sediments with sampling interval of 10 cm from four representative cores LH34, LH40, LH49, and LH56 (**Figure 3**).

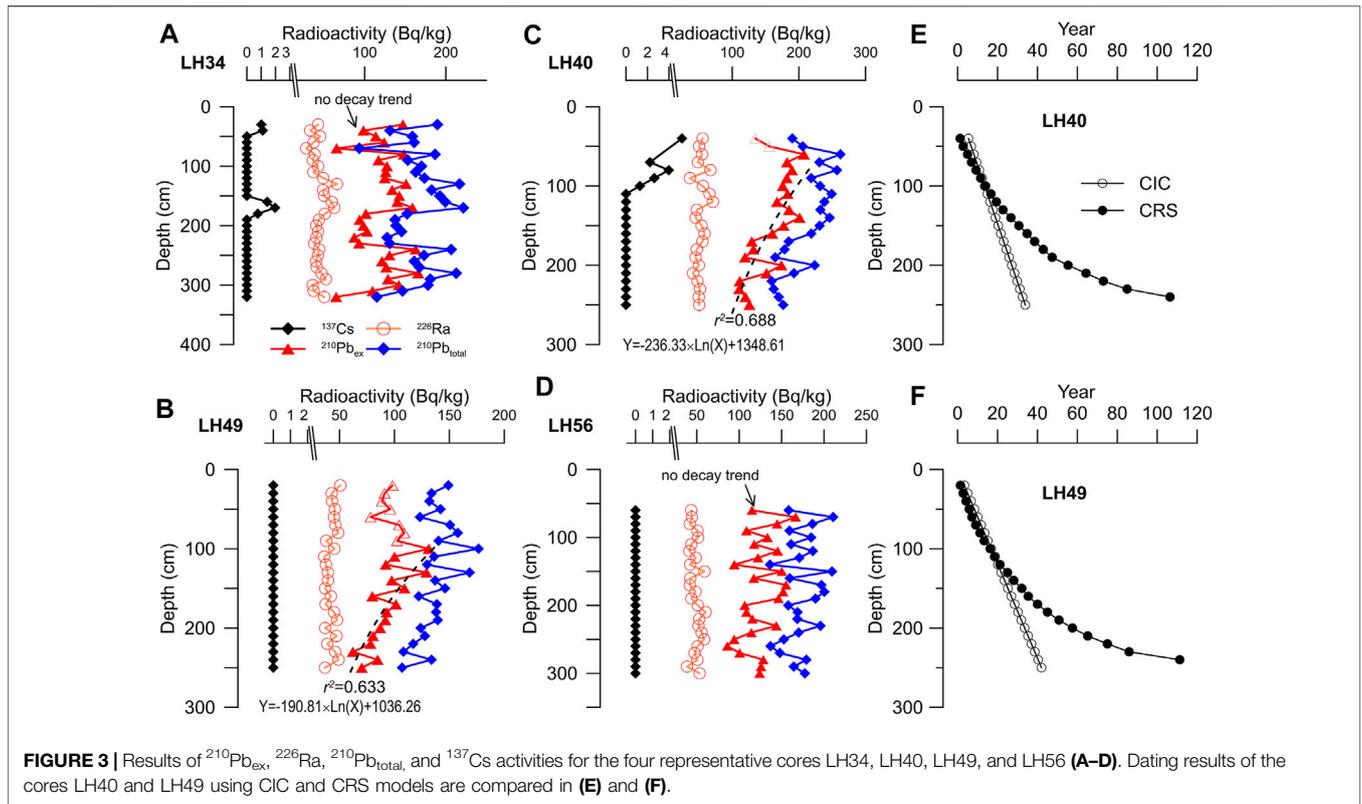


Experiments, Instrumentation, and Data Processing

The materials for ^{210}Pb and ^{137}Cs measurement were first homogenized and grinded. 5–9 g of dried sediments were sealed for 4 weeks to secure equilibrium between radon and its daughters prior to measurement. The activity of total ^{210}Pb , ^{226}Ra , and ^{137}Cs was detected using the high-resolution gamma spectrometer (High purity Ge. EG&G Ortec, the United States) at Nanjing Institute of Geography and Limnology, China Academy of Sciences. As ^{226}Ra activity was assumed to be the same as the supported ^{210}Pb , the concentration of the excess ^{210}Pb ($^{210}\text{Pb}_{\text{ex}}$) was determined by subtracting the total ^{210}Pb activity by ^{226}Ra activity. Two models

with assumptions of the constant rate of supply (CRS) and the constant initial concentration (CIC) were used to establish the ^{210}Pb chronology (Krishnaswamy et al., 1971; Appleby and Oldfield, 1978; Robbins, 1978). The ^{210}Pb disintegration constant of 0.03118/a was employed for the calculation of the sedimentation rate. For the CIC model, the uppermost dots characterized as raised $^{210}\text{Pb}_{\text{ex}}$ activity with the increased depth were excluded for the reconstruction of the age-depth model. For the CRS model, the bulk density of 1.8 g/cm^3 was used for all the dried samples.

XRD analysis of clay mineralogy was conducted on a Bruker D2 Phaser X-ray diffractometer equipped with $\text{CuK}\alpha$ radiation



unit. Chemical treatment was first performed to remove the carbonate and organic matter. The fraction of $<2\ \mu\text{m}$ was separated by setting following Stocks' law. After being concentrated using a centrifuge, the material was smeared on two aliquots, one of which was air-dried, and the other one was mixed with saturated ethylene-glycol over 1 day. XRD measurements were performed with a scanning range of $3\text{--}30^\circ\ 2\theta$. The step is $0.02^\circ\ 2\theta$ with the associated residence time of 0.035 s. Semi-quantitative estimation of peak areas of the basal reflections was conducted for the clay mineral groups of smectite ($15\text{--}17\ \text{\AA}$), illite ($10\ \text{\AA}$), and kaolinite/chlorite ($7\ \text{\AA}$) on the ethylene glycolate curves (Figure 4B). The relative content of kaolinite and chlorite was calculated using the ratio of $3.57/3.54\ \text{\AA}$ peak area.

Non-linear modeling of the clay mineral assemblage in the modern riverine sediments (gray-filled circles in Figure 1B. Liu et al., 2019) was conducted following Yu and Zhang (1998) and Fan et al. (2002). The principle of mass balance of each index was applied in this model to constrain the parameters in the model and to calculate the riverine contributions according to the relative content of the clay minerals in the modern fluvial sediments from the Daling and Liaohe Rivers (Liu et al., 2019). A discriminant was established to distinguish the sediment sources (i.e., the Liaohe or Daling Rivers). The details of the non-linear model are shown as follows.

Supposing there are n source zones, while each source zone has m minerals. The n^{th} source zone sediment concentration percentage is x_n , and the percentage of the m^{th} mineral in the n^{th} source region is a_{nm} . The m^{th} clay mineral content in the core

sediments is b_m . According to the sediment balance principle, the relationship could be listed as

$$\begin{cases} a_{11}x_1 + a_{21}x_2 + \dots + a_{n1}x_n + \varepsilon_1 = b_1 \\ a_{12}x_1 + a_{22}x_2 + \dots + a_{n2}x_n + \varepsilon_2 = b_2 \\ \dots \dots \dots \\ a_{1m}x_1 + a_{2m}x_2 + \dots + a_{nm}x_n + \varepsilon_m = b_m \\ x_1 + x_2 + \dots + x_n \leq 1 \end{cases} \quad (\text{Eq. 1})$$

In addition, the supply amount x_i of each source area should also satisfy the following equations $x_i \geq 0\ (i = 1, 2, \dots, n)$.

The objective function is expressed as follows:

$$\min f(x) = \sum_{i=1}^m \varepsilon_i^2 \quad (\text{Eq. 2})$$

The restrictions are listed as follows:

$$x_i \geq 0\ (i = 1, 2, \dots, n), \quad (\text{Eq. 3})$$

$$\sum_{i=1}^n x_i \leq 1. \quad (\text{Eq. 4})$$

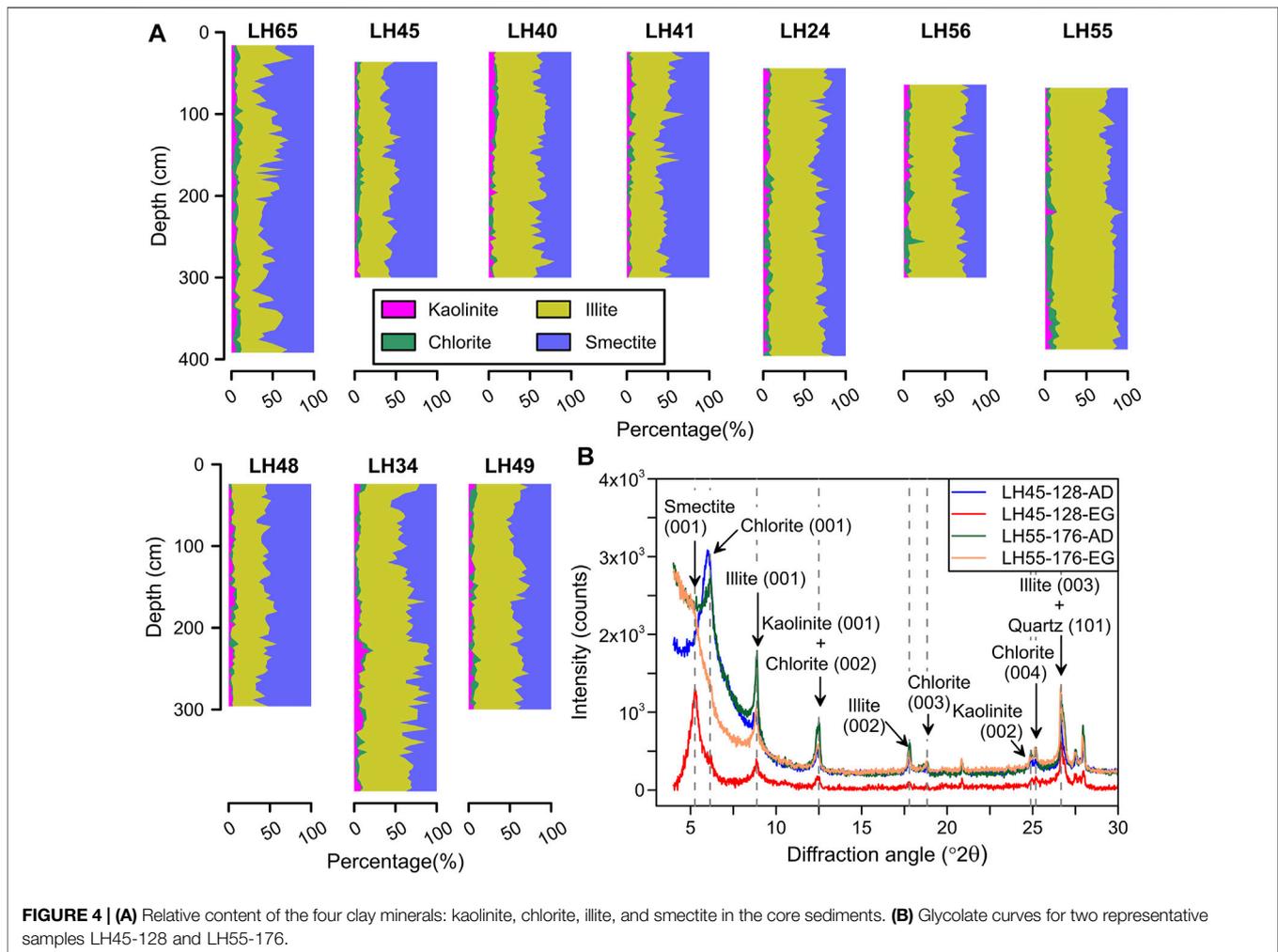
In this study, the sources were Liaohe River (x_1) and Daling River (x_2), and the minerals were smectite (m1), illite (m2), kaolinite (m3), and chlorite (m4). The abovementioned model can be simplified to

$$\min f(x) = \sum_{i=1}^m \varepsilon_i^2 \quad (\text{Eq. 5})$$

while

$$x_1 \geq 0, x_2 \geq 0, \quad (\text{Eq. 6})$$

$$x_1 + x_2 \leq 1. \quad (\text{Eq. 7})$$



The calculation accuracy of the contribution rate of each source area is evaluated. Eq. 1 is written as a matrix, which is $AX + \varepsilon = B$. The variance of B can be written as

$$D_{bb} = E\sigma_0^2, \quad (\text{Eq. 8})$$

where E is the unit matrix and σ_0^2 is the unit weight variance. The covariance matrix of X is

$$D_{xx} = (A^T A)^{-1} A^T D_{bb} A (A^T A)^{-1}, \quad (\text{Eq. 9})$$

$$\sigma_0^2 = \frac{1}{m-n} \varepsilon^T \varepsilon. \quad (\text{Eq. 10})$$

It can be concluded from the above equation that the estimate of the covariance matrix (\widehat{D}_{xx}) of the contribution rate of the source region is

$$\widehat{D}_{xx} = (A^T A)^{-1} A^T A (A^T A)^{-1} \sigma_0^2. \quad (\text{Eq. 11})$$

(\widehat{D}_{xx}) is an n-order symmetric square matrix whose square root is the estimated value of the standard deviations $\sigma_{x1}, \sigma_{x2}, \dots, \sigma_{xn}$ of the contribution ratios x_1, x_2, \dots, x_n (Yu and Zhang, 1998; Fan et al., 2002). The average value of clay minerals in the surface

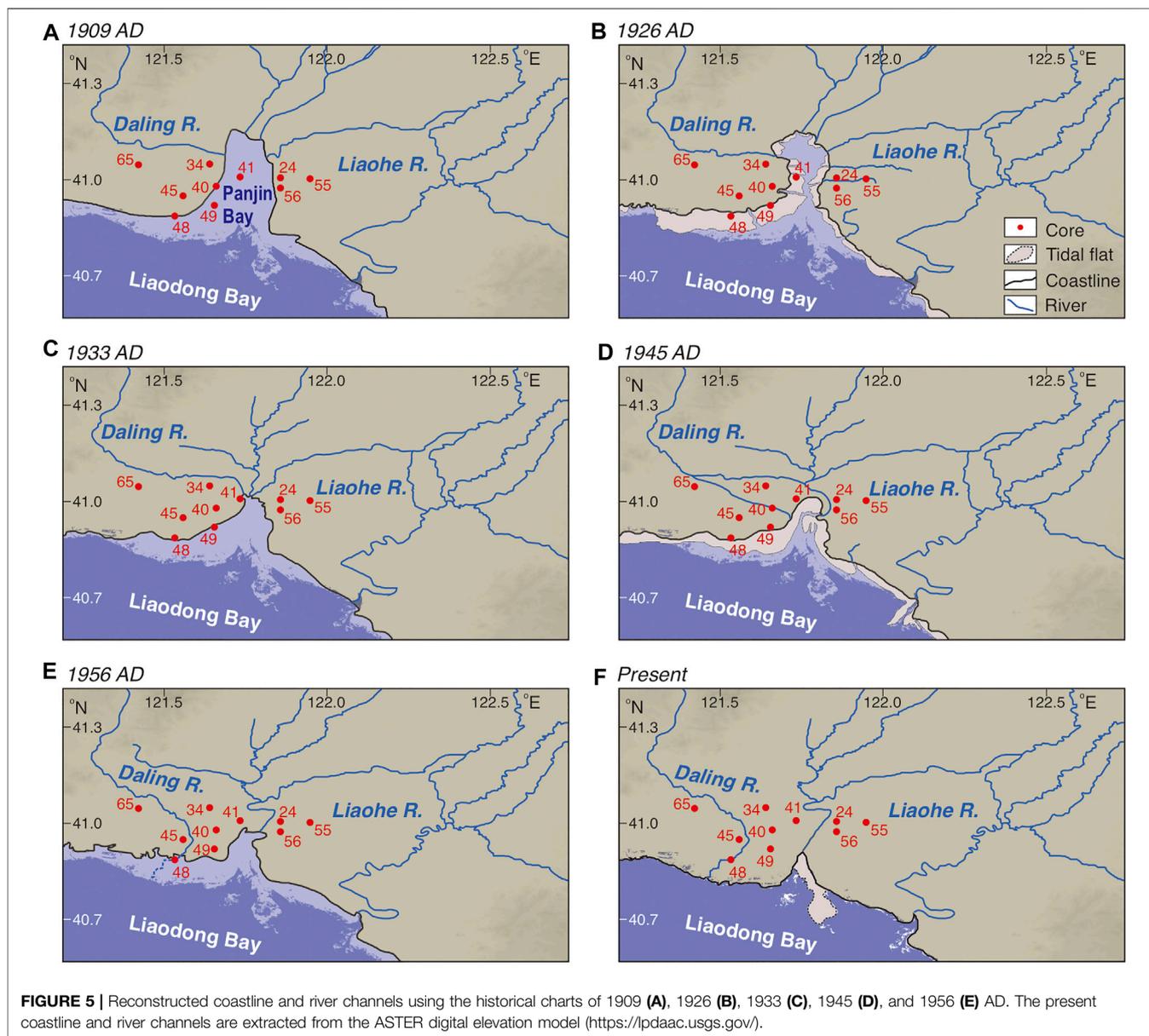
samples of Daling and Liaohe rivers in this study represents the clay mineral characteristics of the two rivers, which is $a_{11} = 28.27\%$, $a_{12} = 58.98\%$, $a_{13} = 6.53\%$, $a_{14} = 6.17\%$; $a_{21} = 58.79\%$, $a_{22} = 34.81\%$, $a_{23} = 3.35\%$, $a_{24} = 3.05\%$. The discriminant was further used to quantify sediment contributions from the Liaohe and Daling rivers in the shallow cores.

Five historical maps and the present satellite image of the NBC were collected to extract information about the paleo-coastlines and river channels in the years 1909 AD, 1926 AD, 1933 AD, 1945 AD, 1956 AD, and the present (Figure 5). All the historical maps were converted to the 1980 Xi'an coordinate system and georectified using the locations of historical towns. The paleo-coastline and river channel were subsequently digitalized and compared with those extracted from the modern imagery (Figure 5).

RESULTS

^{210}Pb and ^{137}Cs Dating Results

$^{210}\text{Pb}_{\text{ex}}$ and ^{137}Cs activities of the sediments in cores LH34, LH40, LH49, and LH56 are shown in Figure 3. $^{210}\text{Pb}_{\text{ex}}$ activities overall vary between 100 and 200 Bq/kg. ^{226}Ra activities are generally



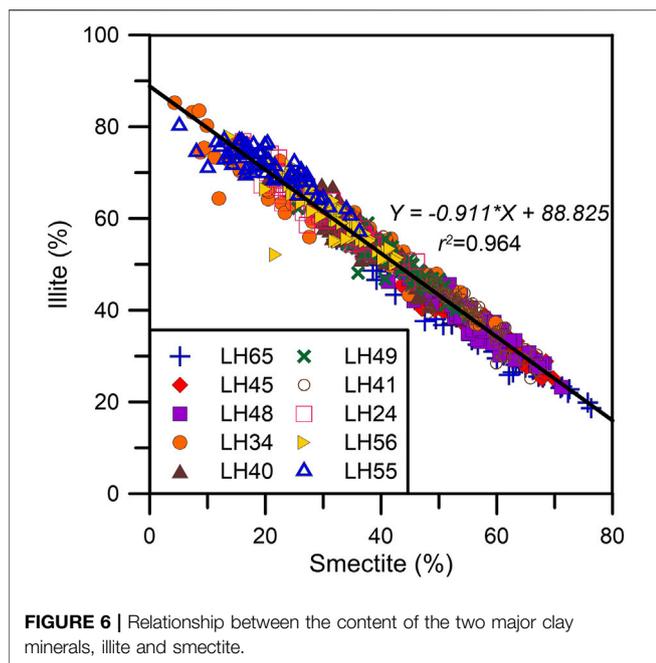
constant for the sediments in each core. $^{210}\text{Pb}_{\text{ex}}$ and $^{210}\text{Pb}_{\text{total}}$ activities in cores LH40 and LH49 show a common decay with an increase in depth, which is yet not observed in cores LH34 and LH56 (Figures 3A–D). The upper layer with increased $^{210}\text{Pb}_{\text{ex}}$ activity downward in cores LH40 and LH49 is regarded as a disturbed layer, which is thus excluded for age determination. Two models were applied for curve fitting and sedimentation rate (SR) calculation. For the CIC model, the $^{210}\text{Pb}_{\text{ex}}$ activity–depth plot was fitted following logarithmic decay (Krishnaswamy et al., 1971). The mean SRs of 7.37 and 5.95 cm/a were determined for cores LH40 and LH49, for which the coefficient-of-determination (r^2) are 0.688 and 0.633, respectively. According to the fitting results using the CIC model, the sediments for cores LH40 and LH49 dated back to the 1980s and 1970s, respectively. For the CRS model, the initial concentration for each core was first calculated. Timing and SR of

each layer were then determined (Appleby and Oldfield, 1978; Robbins, 1978). The timing of the sedimentation for cores LH40 and LH49 is ~ 100 years (Figures 3E,F).

Compared to $^{210}\text{Pb}_{\text{ex}}$, ^{137}Cs peaks were observed in cores LH34 and LH40 with much lower activity of $<ca. 4$ Bq/kg, while it was too low to be detected for the core sediments in LH49 and LH56. The low activity of ^{137}Cs might induce large uncertainty, so that attempt of constraining the timing of sedimentation in the NBC using the ^{137}Cs dating approach was abandoned in this study.

Clay Mineral Compositions of the Core Sediments

Clay mineral compositions for the sediments in the ten representative cores are shown in Figure 4A. Illite (18.6–85.3%,



average 51.5%) and smectite (4.3–76.3, average 41.0%) are the major clay minerals in the NBC, while the content of kaolinite (0–19.7%, average 3.8%) and chlorite (0–25.9%, average 3.7%) is minor. It agrees with the outcome of Liu et al. (2019), as illite and smectite are the dominant fractions for the modern riverine sediments in the NBC, while the content of chlorite and kaolinite is generally lower than that of *ca.* 10%. The relative content of illite and smectite shows a negative linear relationship (Figure 6).

The content of smectite overall decreases from west to east (Figure 4). In vertical, it shows a decreasing upward in the lower part and an increasing upward in the upper part of the cores in the west sub-zone, whilst it demonstrates a gradual increasing upward in the lower part and a decreasing upward in the upper part of the cores to the east. The content of smectite in the eastmost core LH55, at a relatively low level, generally increases upwards. According to the negative linear relationship between assemblages of smectite and illite (Figure 6), the composition of illite varies oppositely in spatial and vertical to the smectite composition. Compared to smectite and illite, relatively minor kaolinite and chlorite are contained in all the core sediments, which do not show obvious variation tendency spatially and vertically (Figure 4).

The Daling riverine sediments tend to contain more smectite than illite, whilst illite is the dominant fraction in the Liaohe originated sediments (Liu et al., 2019). The amounts of the clay mineral groups kaolinite + chlorite, illite, and smectite for the riverine and core sediments are thus plotted in the ternary diagram for preliminary sediment provenance discrimination (Dou et al., 2014; Liu et al., 2019, Figure 7). The clay mineral assemblages for the core sediments in the three sub-zones were plotted in Figures 7B–D. It clearly demonstrates that sediments in the west and east sub-zones provide similar clay mineral

compositions to the Daling and Liaohe modern riverine sediments, respectively, whilst in the middle sub-zone mixture of the sediments of different sources is revealed, attributing to contribution from both the two rivers. Clay mineral assemblages show that some sediments in the west sub-zone are characterized as the Liaohe River input, which is likely related to river channel migration and transportation by the longshore current.

Quantification of River Input by Non-Linear Modeling

The content of the four clay minerals in the modern riverine sediments was used for non-linear discriminant analysis of sediment provenance. The assumption is that the Daling and Liaohe rivers are the two sources of the sediments in the NBC over the last century. The constructed discriminant was then applied to quantify the contribution from the Liaohe and Daling rivers for the core sediments. The contribution ratios of the two rivers are shown in Figure 8. It shows that over 90% of the sediments deposited in the eastern area of the NBC were derived from the Liaohe River, and the rest sediments were transported by the Daling River. The sediments deposited in the western area were mainly derived from the Daling River with a small amount of Liaohe fluvial input (Figure 8) and those in the middle area of the NBC were supplied from both, which are regarded as the major and secondary sediment provenances, respectively. In the upper section of the three cores in CC', sediments of Daling River origin, varying from 0.01 to 98.92%, tend to increase seawards. It is noticed that sediments in core LH41, close to the modern Liaohe River channel, are discriminated to be supplied mainly by the Daling River (Figure 8).

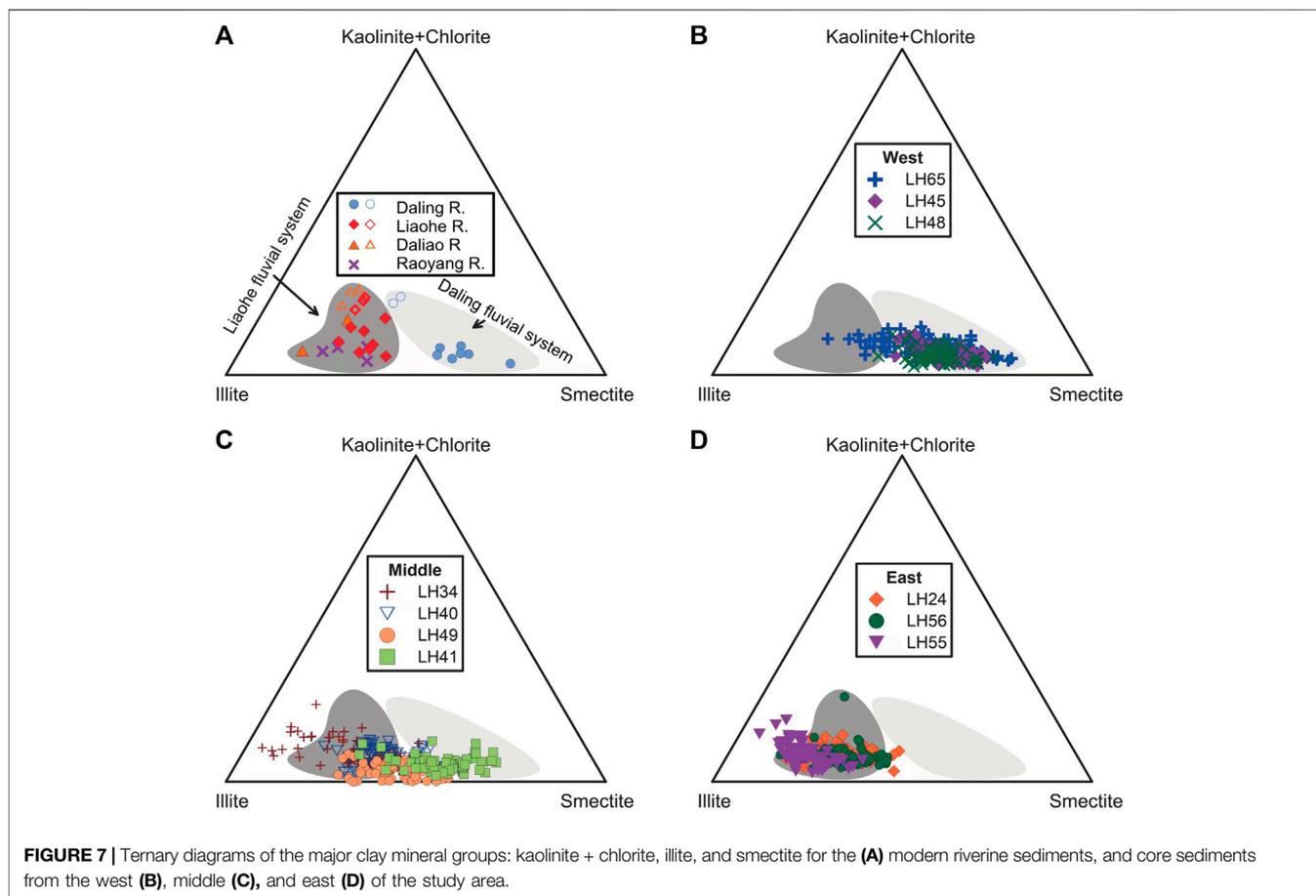
Coastline and River Channel Migrations in the NBC Since the Last Century

The information on the coastline and river channel migration was extracted from six images (Figure 5). The so-called 'Panjin Bay' was infilled gradually due to progradation during the last century. The Liaohe River and local tributaries generally flow south- and southwest-wards to the Panjin Bay, whilst the channel of the Daling River changed significantly in the last century. Before 1933 AD, the Daling River channel flew eastwards, while the tributary of the Daling River joined those of the Liaohe River and flew into the Panjin Bay westwards. After 1956 AD, the Daling River flows into the Liaodong Bay southwards (Figure 5).

DISCUSSION

Sedimentation Rate (SR) in the North Bohai Coast

The CIC and CRS modeling results for the sediments in cores LH40 and LH49 show a clear discrepancy, as the mean, SRs determined using the CIC model are much larger than those by CRS modeling (Figures 3E,F). Reliability of the SR by ^{210}Pb dating for the two cores could be assessed by down-core lithological variation and historical coastline migration. It demonstrates that sediment color transition from brownish-gray to light brown upwards in vertical resulted from



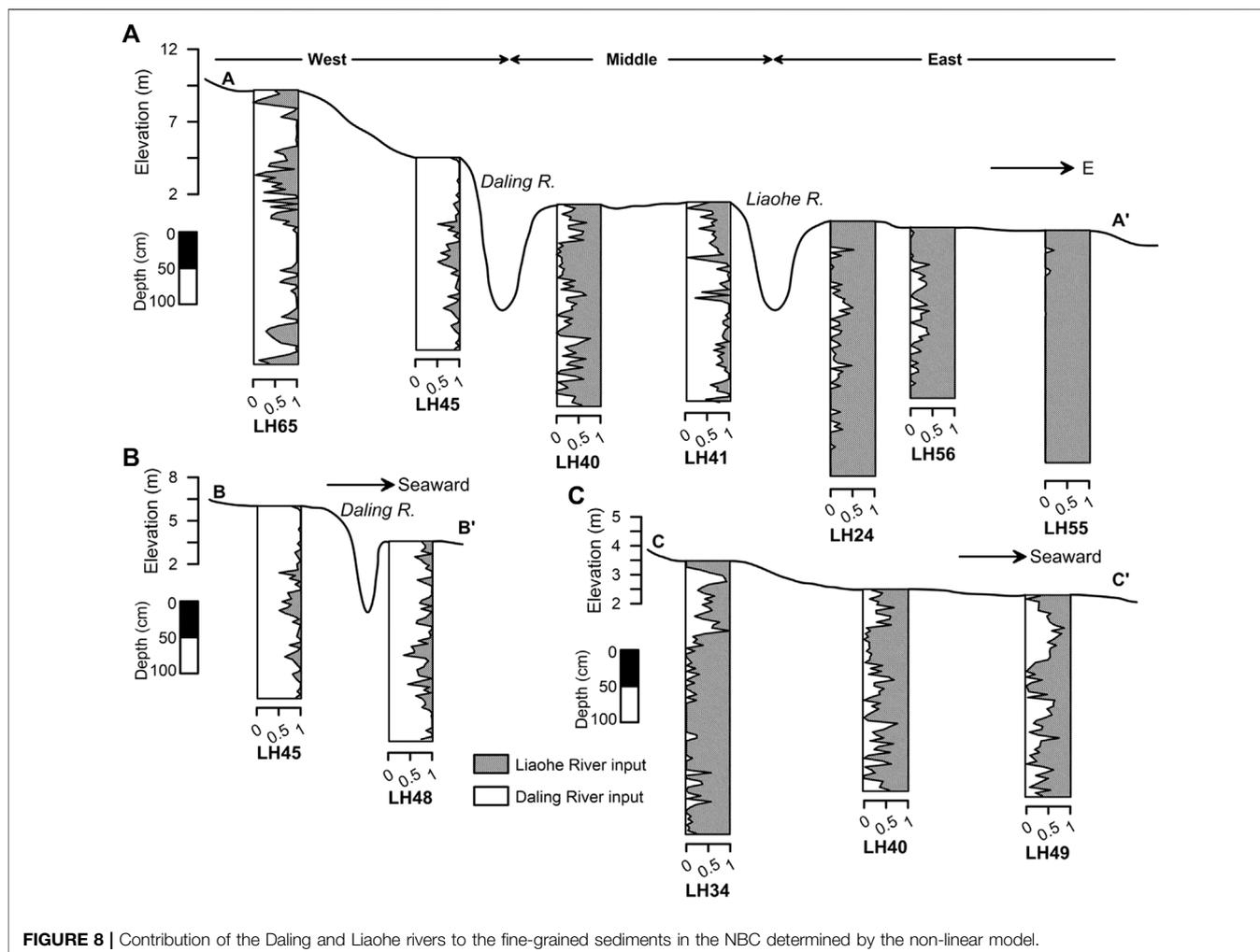
sedimentary environment change (Figure 2), which was directly induced by the seaward retreat of the coastline. Core LH40 is located between the coastlines of the years 1909 and 1926 AD (Figures 5A,B), for which the boundary of sediment color change is at depth of 205 cm (Figure 2). The mean SR of ca. 1.9 cm/a is thus determined for the upper succession of core LH40 accordingly. The depth of color variation boundary is 180 cm for core LH49 (Figure 2), for which the location is between the coastlines of 1933 and 1945 AD (Figures 5C,D). The mean SR is ca. 2.0–2.3 cm/a. Based on the constraints from lithologic characteristics and coastline migrations, the CIC model yielded SRs larger than 5 cm/a are proven unreliable.

With the mean SRs close to 2.0 cm/a, the results after CRS modeling are more satisfactory compared to that using the CIC model. The timing of the marine-terrestrial transition is 1955 AD for core LH40 and 1970 AD for core LH49 when using the CRS chronological framework for each layer. It implies that the SRs for the upper succession under the terrestrial environment is still overestimated. However, if the mean SR for the whole core was used, the timings of marine-terrestrial transition are 1927 and 1934 AD for cores LH40 and LH49, respectively, which are in general consistent with the age constraints by lithology and geomorphology.

SR overestimation for ^{210}Pb dating is also evidenced by the previously published dating results using optically stimulated

luminescence (OSL) and accelerator mass spectrometry radiocarbon-14 (AMS^{14}C) dating approaches. Numerical ages and depths of the uppermost chronological samples in the cores in the NBC were used to calculate the mean SRs in the last few centuries (Li et al., 2014; Ma et al., 2016; He et al., 2018; Li Y. et al., 2018). It shows that the SRs were generally smaller than 2 cm/a in the NBC in the late Holocene. Furthermore, ^{210}Pb and ^{137}Cs dating approaches have been successfully conducted on the sediments in the NBC by Li H. et al. (2019), which suggested that the SR was ca. 1.6–2.7 cm/a for the studied cores sediments. SR overestimation of ^{210}Pb chronology is likely attributed to the un-negligible external input of ^{210}Pb due to the complex hydrodynamics in the coastal area (Wang et al., 2016). It results in either an unclear decay trend of $^{210}\text{Pb}_{\text{ex}}$ activity (i.e. cores LH34 and LH56) or overestimated SRs using inappropriate age model (i.e. cores LH40 and LH49) in this study (Figure 3).

Overall, by comparing the ^{210}Pb dating results with the lithological constraints and other results by numerical dating, ^{210}Pb dating using the CIC model failed to constrain the timing of the sedimentation in the NBC over the last century in this study. This is probably due to the unstable sedimentary environment and complex marine-terrestrial exchange in the coastal area. CRS model seems to yield slightly overestimated SR, as the SR increased over the last century, which is in contradiction with the declined fluvial sediment flux. However, the mean SR appears to be reliable, for

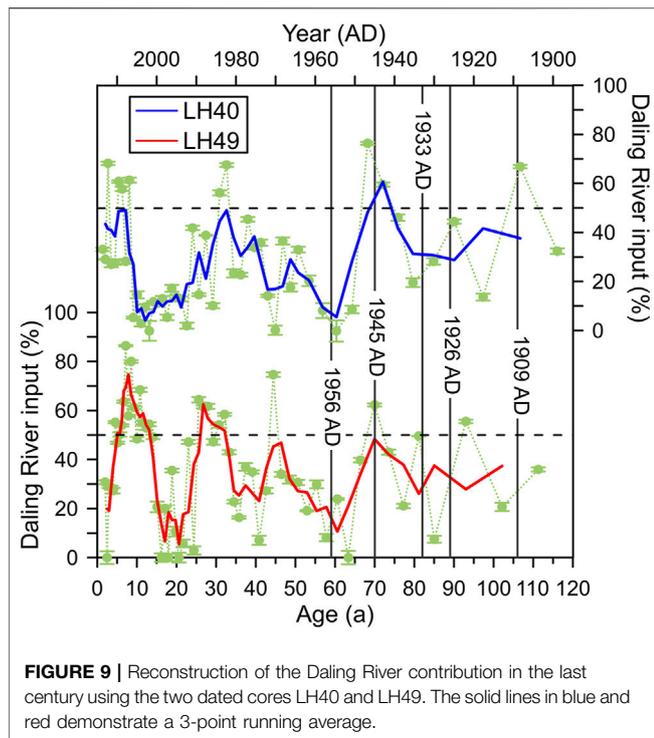


which the timing of the marine-terrestrial transitional boundary agrees with that constrained by the history of coastline retreats.

Clay Mineral Sources and Its Morphological Implication

The NBC receives sediments from the Liaohe and Daling rivers, which stretch across the monsoon and non-monsoon regions in northern China, where the climate condition of the drainage basins possibly induces physical erosion of the bedrocks. Smectite and illite are the two major clay minerals contained in the sediments in the NBC, which correlate to the Daling and Liaohe rivers as the two end-members of the sediment sources (Liu et al., 2019). Limited by the range of the drainage basin, it is clear that the core sediments collected from the east NBC correspond to major contribution of the Liaohe River, while those from the west NBC are mainly derived from the Daling River (Figures 7B,D). In contrast, the clay minerals in the core sediments were derived from both the two rivers in the middle zone of the study area (Figure 7C), which were induced by fluvial and coastal morphologic evolution over the last century.

The coastal morphological evolution in the NBC over the last century was reconstructed using the two dated cores and the adjacent cores, in conjunction with the information on the historical coastline and river channel migration. Based on the ^{210}Pb chronology after CRS modeling, variation of the river input is shown in Figure 9. It demonstrates that clay minerals in the two cores have documented consistent variation of the fluvial contribution. The Panjin Bay was infilled by the sediments from the two rivers since 1909 AD, which resulted in significant coastal morphological change. During this period, the Daling River flew eastward, while the Liaohe and its local tributaries flew into the Panjin Bay south- and westwards (Figure 5). Transported by fluvial input and the coastal current in the Panjin Bay, clay minerals from the offshore cores LH40, LH41, and LH49 showed a mixture of the Daling and Liaohe River sources. With further infilling of the Panjin Bay, the coastline in the west Panjin Bay retreated seawards significantly, resulting in an increase of the Daling River input during 1909–1926 AD (Figure 9). It is also noticed that sediments in the cores closer to the Daling estuary responded earlier. From 1926 to 1933 AD, the north coast of the Panjin Bay was rapidly filled mainly by sediments of



Liaohe origin after comparing the coastline migration during the period (Figures 5B,C). Consistently, the relative contribution of the Daling River declined in cores LH40 and LH49 according to the non-linear modeling. The river channel of the Daling River migrated during 1933–1945 AD southeastward, which induced increasing river input to the area where cores LH40 and LH49 were located. Based on the outcome of provenance discrimination, the migration was relatively rapid, as the response in the two cores was possibly simultaneous (Figure 9). Since the Daling River further flew to the Liaodong Bay southward, the influence on the area of cores LH40 and LH49 from the Daling River became weaker, which correlates to an increase in Liaohe contribution as revealed by the modeled provenance quantification. In summary, with a comparison between the historical morphological change and the outcome of provenance discrimination, the Daling River input was major in the 1910s and 1940s, which attributed to the rapid progradation and river channel migration, respectively. Although the tendency of fluvial contribution in different cores is in agreement, it is shown that the fluvial contribution of clay minerals is related to the relative position between the estuary and the cores. During 1909–1945 AD, the Daling estuary was close to the location of core LH40, leading to more fluvial contribution in core LH40 from the Daling river. On the contrary, the Daling estuary migrated southward after 1945 AD, which was closer to core LH49. This resulted in more Daling River input in core LH49 compared to core LH40.

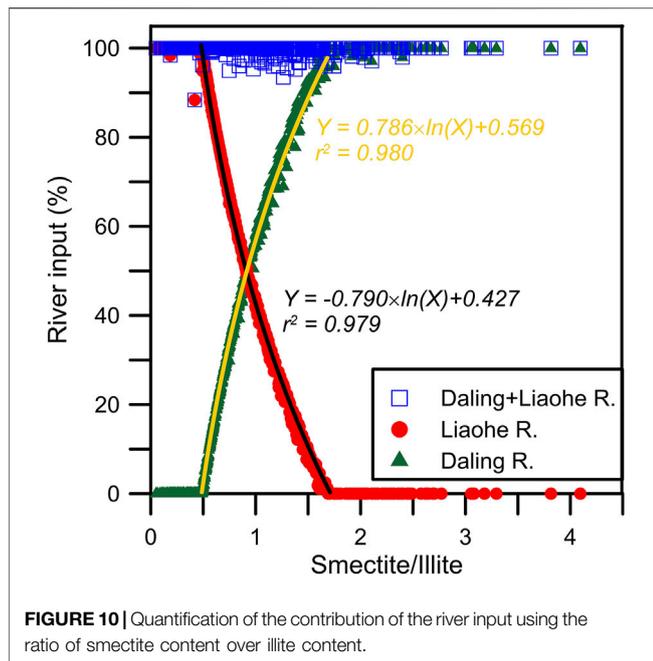
The consistency between the non-linear modeling results, the river channel, and coastal migration has been also reflected

by the adjacent cores without direct dating results. For instance, non-linear modeling of the provenance discrimination shows that the fine-grained sediments in core LH41, that is close to the modern Liaohe River channel (Figure 1), originated mainly from the Daling River (Figure 8). Based on the information on the historical river channel, the Daling River flew into the Panjin Bay southeastwards before 1956 AD, resulting in progradation southeastward and infilling of the Panjin Bay between 1909–1956 AD (Figure 5). The majority of sediments in core LH41 of the Daling riverine contribution attributes to its location in the estuary of the Daling River during 1909–1956 AD. It is also noticed that the tributary of the Daling River joined those of the Liaohe River and flew into the Panjin Bay westwards revealed by the historical map in 1945 AD (Figure 5D). It resulted in minor input from the Daling River on the east coast of Panjin Bay (LH24 and LH56. Figure 8). After 1956 AD, the relative contribution of the Daling River increased at least twice. i.e., in the 1980s and 2000s (Figure 9). It might be related to flood events, e.g., the flood in the year of 1984, or anthropogenic impact, which needs more detailed investigation. Overall, it indicates that the determination of the riverine input for the two rivers after non-linear modeling is consistent with the reconstructed historical river channel and coastline migration.

Potential Applicability for Paleoenvironmental Studies in the North Bohai Coast

The NBC is one of the depocenters in the Bohai Sea. The thick sediments contain rich paleoenvironmental information (Liaoning Geological Bureau, 1983; IOCAS, 1985). According to the discussion abovementioned, clay mineralogy is useful to discriminate the provenance and reconstruct morphological evolution. Meanwhile, clay minerals as one of the applicable environmental proxies could be also used to investigate the paleoclimate change and sediment transportation process (e.g., Liu et al., 2003; Li J. et al., 2018; Yu et al., 2019), which is, however, has not been used to interpret paleoenvironment change in the NBC.

The variation of the clay mineral assemblages is not correlated to paleoclimatic change/chemical weathering conditions straightforwardly (Thiry, 2000; Liu et al., 2003). For the purpose of paleoenvironmental investigation using clay mineralogy in the NBC, it is essential to separate the sediments with a single origin from those with mixed sources. To simplify the non-linear model in this study, the contributions of Daling and Liaohe rivers were presented as the content ratio of smectite/illite. The ratio was plotted against the determined contribution of the two rivers (Figure 10). Sediments most likely derive from the Liaohe River when the smectite/illite ratio is lower than 0.5, while those are solely associated with Daling River input when the ratio is higher than 1.7. A logarithm correlation of smectite/illite ratio with the percentage of river input for both the rivers is observed for the ratio range between 0.5–1.7. The correlation with fitting equations shown in Figure 10 can be potentially applied to quantitatively discriminate the contributions of fluvial inputs for the quaternary



provenance-unknown sediments in the NBC, and further extract the clay mineral content derived from the signal provenance.

CONCLUSION

In this study, the clay mineralogy of the coastal sediments in the North Bohai Coast (NBC) was investigated to determine the variation of the fine-grained sediment sources over the last century. Clay mineral content in the modern riverine sediments in the NBC was used to discriminate the sediment provenance and to quantify the sedimentary contribution from different fluvial systems. ^{210}Pb and ^{137}Cs radioactivity was measured for the dating attempt. The migration of coastline and river channel in the North Bohai Coast during the last hundred year was reconstructed by digitalizing the historical charts and satellite images. The main conclusions are drawn as follows:

- ^{210}Pb chronology after CIC modeling overestimates the sedimentation rate, but the CRS model results in an acceptable sedimentation rate for two dated cores. ^{137}Cs dating approach is not applicable due to the low activity of the nuclide.
- Contribution of the Daling and Liaohe Rivers has been quantified using non-linear modeling of the clay mineral contents. The modeling results demonstrate a satisfactory relationship with the smectite/illite content ratio, which is also consistent with the lithological and coastal geomorphologic constraints.

- Clay minerals in western and eastern NBC are mainly derived from the Daling and Liaohe Rivers, respectively. In the middle NBC, the Daling River input was relatively major during the 1910s, 1940s, 1980s, and 2000s. The increase of Daling River input in the first two periods resulted from progradation and river channel migration, respectively.
- Increasing fluvial contribution from the Daling River in 1980s and 2000s might be related to flooding events and/or anthropogenic impact, which needs more investigations. The fluvial contribution from the Liaohe River was major in the middle NBC during 1920–1940 AD, likely attributing to rapid progradation. River channel migration of the Daling River after 1956 AD likely resulted in a relative increase of Liaohe River input during 1950–1970 AD, as the influence on the middle NBC from the Daling River was weaker.

DATA AVAILABILITY STATEMENT

The raw data supporting the conclusion of this article will be made available by the authors, without undue reservation.

AUTHOR CONTRIBUTIONS

Conceptualization and methodology, YL and DL; formal analysis, SS, DL, KH, and YL; original draft preparation, YL, DL and SS; and review and editing, KH and YL. All authors have read and agreed to the published version of the manuscript.

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