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Differences in radiocarbon ages among molluscan shells, plant materials, and total organic carbon: An example from the paleo-Changjiang incised-valley fill, China

Kazuaki Hori ^{a, *}, Yoshiaki Saito ^{b, c, **}^a Department of Geography, Graduate School of Environmental Studies, Nagoya University, Nagoya 464-8601, Japan^b Geological Survey of Japan, AIST, Tsukuba 305-8567, Japan^c Estuary Research Center, Shimane University, Nishikawatsu-cho 1060, Matsue 690-8504, Japan

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ABSTRACT

Radiocarbon dating of samples from the CM97 cores of Late Pleistocene to Holocene sediments in the paleo-Changjiang incised valley, China, was performed, and age differences among molluscan shells, wood and other plant materials, and bulk total organic carbon (TOC) were evaluated. The incised-valley fill consists of fluvial, estuarine, and deltaic deposits in ascending order. In the dating results, samples of wood and other plant materials yielded similar ages to molluscan shell samples. In contrast, in samples from estuarine and deltaic sediments, radiocarbon ages of TOC were systematically older, by up to 6500 calibrated years, than those of molluscan shells. This result suggests that the paleo-Changjiang River transported aged terrigenous organic matter to its mouth. The magnitude of the age offset increased upward within the transgressive estuarine sediments, and the largest offset, ~4500 calibrated years on average, was observed in the prodelta sediments deposited after the maximum transgression. Grain size, the C/N ratio, $\delta^{13}\text{C}$ values, and the sediment accumulation rate of the incised-valley-fill sediments and the paleo-water depth indicated a weak terrestrial influence and a strong marine influence in the prodelta sediments. Therefore, the large age offsets in the prodelta sediments likely reflect reworking and resuspension in a strongly tide-dominated setting of sediments deposited earlier on the inner shelf, resulting in the supply of both fine-grained resuspended sediments and old organic carbon to the prodelta in addition to the supply of terrigenous organic carbon from the paleo-Changjiang River. The upward (deepening) increase of the age offset in the estuarine sediments supports this interpretation. The magnitude of the age offset in the past prodelta sediments was comparable to the old radiocarbon ages of the surface sediments in the present prodelta to inner shelf area. This result suggests that the supply, dispersal, recycling, and burial of terrestrial organic carbon on the inner shelf has been mostly stable during the past 8000 years.

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1. Introduction

Radiocarbon dating by accelerator mass spectrometry is a widely used dating method that can be applied to very small samples. Thus, even if there are no carbonate (e.g., molluscan shells, foraminiferal tests) or plant materials (e.g., wood, seeds) in

sediments, it is possible to measure radiocarbon ages on organic carbon extracted from the sediments (e.g., Ikehara, 2000; Ohkouchi et al., 2002, 2003). Comparisons of radiocarbon ages between bulk total organic carbon (TOC) and other materials from core sediments obtained from various environments, including estuaries (Colman et al., 2002), coastal lowlands (Ishimura et al., 2016), and forearc and backarc basins (Ikehara, 2000) have shown that bulk TOC samples tend to be systematically older than carbonate samples and, thus, unreliable, probably because of the input of aged, terrigenous organic carbon to the receiving basins. Compound-specific radiocarbon analysis (Eglinton et al., 1996) of marine sediments (Eglinton et al., 1997; Pearson and Eglinton, 2000; Uchida

* Corresponding author.

** Corresponding author. Geological Survey of Japan, AIST, Tsukuba 305-8567, Japan.

E-mail addresses: khori@lit.nagoya-u.ac.jp (K. Hori), ysaito@soc.shimane-u.ac.jp (Y. Saito).

et al., 2000; Ohkouchi et al., 2002) has provided more reliable ages of organic carbon as well as important information about present and past carbon cycles.

Sedimentary environment (e.g., terrigenous sediment input, primary production, water depth, and salinity) has undergone great changes especially in the present river mouth and the adjacent continental shelf with the influence of climate and sea-level changes since the Last Glacial Maximum (LGM). Thus, the magnitude of age offsets between carbonate and bulk TOC samples obtained from coastal areas may display temporal variation rather than stable value. Incised-valley fill deposits, which record fluvial, estuarine, and delta environment in response to a sea-level rise and the following sea level highstand, are suitable for verifying whether the temporal variation has occurred.

Raymond and Bauer (2001) showed by the measurement of ^{14}C and ^{13}C in water samples that the Amazon River and some North American rivers flowing into the North Atlantic Ocean transport old terrestrial particulate organic carbon (POC). $\Delta^{14}\text{C}$ values of POC in the rivers are significantly depleted compared to dissolved organic carbon and indicate ages of hundreds to thousands of years. The Huanghe (Yellow) and Changjiang (Yangtze) rivers, the two largest rivers in China, which flow into the Bohai Sea and the East China Sea, respectively, also carry POC ranging from in age from 800 to 8000 ^{14}C years (Wang et al., 2012; Tao et al., 2015). The nature, relative proportions, sources, and burial processes of terrestrial and marine organic matter preserved in river-dominated continental shelf sediments, such as in the Gulf of Mexico and the East China Sea, have been investigated by various geochemical analyses (Goñi et al., 1997, 1998; Wu et al., 2013; Bao et al., 2016). For example, Wu et al. (2013) reported that sedimentary organic matter around the present Changjiang River mouth (inner shelf region) consists dominantly of aged ($\Delta^{14}\text{C} = -423 \pm 42\%$) organic matter associated with fine-grained sediments. This finding suggests that sedimentary organic matter on the inner shelf has been strongly influenced by inputs of aged organic carbon from the Changjiang River. However, it is unknown whether the input of aged organic matter continues to occur or to what extent it affected past environments in this region. Radiocarbon ages of TOC from sediments from long borehole cores obtained near present-day river mouths may provide information about past inputs of aged organic carbon from these rivers.

In this study, we determined radiocarbon ages on samples of molluscan shells, wood and other plant materials, and bulk TOC from the paleo-Changjiang incised-valley fill, which records depositional environmental changes of river mouth and the adjacent continental shelf in response to sea-level changes during the past 11,000 ^{14}C years (Hori et al., 2002). We then evaluated reliability of these ages to construct an accurate age–elevation model for incised-valley fills in relation to the sedimentary facies and geochemical properties of the deposits. Finally, we discussed the meaning of the magnitude of age offsets between molluscan shells and bulk TOC and its temporal variability during the Holocene by considering depositional environmental change and source and fate of organic carbon.

2. Study area

The Changjiang River begins from glaciers on the eastern Tibetan Plateau and flows 6300 km eastward to the East China Sea. The drainage basin area is approximately $1.8 \times 10^6 \text{ km}^2$. The river is the fifth largest in terms of water discharge ($920 \text{ km}^3/\text{yr}$) and, historically, the fourth largest in terms of its suspended sediment load ($480 \times 10^6 \text{ t/yr}$) (Milliman and Syvitski, 1992). However, the sediment load has decreased by more than 50% since the 1950s and 1960s, because of the construction of numerous dams and

reservoirs as well as the implementation of a water-soil conservation plan in the basin, whereas the water discharge has increased slightly (Yang et al., 2006; Wang et al., 2011).

The present mean tidal range near the river mouth is 2.7 m, and the maximum tidal range approaches 4.6 m (Shen et al., 1988). The tidal current reaches 290 km upstream from the mouth during the dry season, but only 180 km upstream during the flood season (Shen, 1999). The present mean and maximum wave heights within the mouth are 0.9 and 6.2 m, respectively (Zhu et al., 1988).

The river has formed a large tide-dominated muddy delta (Orton and Reading, 1993) with a funnel-shaped morphology and several distributary channels (Fig. 1). The apex of the Holocene Changjiang delta is more than 250 km upstream from the river's mouth, around Yangzhou and Zhenjiang cities. River-mouth sand bars of the present delta are elongated and almost parallel to the river channels. Extensive intertidal flats are developed along the eastern coast of the delta plain.

The subaqueous delta is divided into the delta front and prodelta. Moreover, the delta front can be subdivided into the delta-front platform and the delta-front slope; the platform, which is at water depths of less than 5–10 m, is relatively flat with sand shoals, and the slope extends from 5 to 10 m to approximately 15–30 m in water depth. The prodelta is at water depths greater than 15–30 m. The grain size of the seafloor sediments decreases from delta front to prodelta (Fig. 1).

The paleo-Changjiang incised valley was formed beneath the main part of the present delta plain during the sea-level fall and subsequent lowstand, around the time of the LGM. The paleo-valley merges with the present valley topography on the East China Sea continental shelf (Fig. 1). The incised valley is up to 70 km wide, and its thalweg has a maximum depth of about 80–90 m below the present-day sea level at the river mouth. The valley is filled with fluvial, estuarine, and deltaic sediments in ascending stratigraphic order, deposited mainly during the post-LGM sea-level rise and Holocene highstand. The coastline at the time of the maximum Holocene transgression, ~7000 yr BP, was near Yangzhou (Fig. 1) (Hori et al., 2002; Song et al., 2013).

3. Materials and methods

Three borehole cores, CM97-A, -B, and -C, were obtained from Chongming Island, part of the present delta plain overlying the paleo-Changjiang incised valley in 1997 (Hori et al., 2001a, b) (Fig. 1: site CM97, Holes A, B, and C, latitude $31^\circ 37' \text{N}$, longitude $121^\circ 23' \text{E}$). The three holes are in very close proximity, within ~10 m of one another. Core CM97-A was recovered from 1.60 to 11.75 m depth below the ground level, CM97-B from 8.55 to 70.50 m depth, and core CM97-C from 2.0 to 61.50 m depth. The three core sites range in elevation between 2.38 and 2.48 m above mean sea level.

The sediment facies were analyzed and the sand and mud content were measured in the CM97-A and -B cores. The mud content (<0.063 mm) was measured in 5-cm-thick samples collected every 20 cm (Hori et al., 2001a, b). Kuramoto (2000) measured TOC, $\delta^{13}\text{C}$, and the TOC/total nitrogen ratio (hereinafter, the C/N ratio) in bulk sediments from the CM97-A and -B cores (Fig. 2).

Radiocarbon ages on molluscan shells were determined and reported previously (Hori et al., 2001a). For this study, radiocarbon ages were measured on wood and other plant materials, and bulk TOC extracted from the mud deposits of each core. All ages were measured by accelerator mass spectrometry by Beta Analytic Inc. (Table 1). Shells were pretreated with acid etches. Acid-alkali-acid and acid washes were performed for wood and other plant materials, and bulk sediment samples, respectively.

Calendar ages were obtained by using OxCal v4.2.4 software

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