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Baseline

Dual carbon isotope ($\delta^{13}C$ and $\Delta^{14}C$) characterization of particulate organic carbon in the Geum and Seomjin estuaries, South Korea

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ABSTRACT

We investigated the source, composition, and reactivity of particulate organic carbon (POC) in two contrasting Korean estuary systems, a closed estuary (Geum) (i.e., with an estuary dam at the river mouth) and an open (Seomjin) estuary. A dual isotope ($\delta^{13}C_{POC}$ and $\Delta^{14}C_{POC}$) approach was applied to surface water samples collected along a salinity gradient in August 2016. Our results indicate that phytoplankton-derived POC was the main contributor to the total POC pool in the reservoir of the Geum estuary, while terrestrial-derived POC predominated the upper Seomjin estuary. A simple binary mixing model using $\Delta^{14}C_{POC}$ revealed a higher modern POC contribution (87–90%) in the Geum estuary reservoir than that (77%) of the upper Seomjin estuary. Accordingly, it appears that an estuary dam can alter the source and reactivity of POC in a reservoir, which can be transferred to the adjacent coastal ecosystem.

Estuaries play a crucial role in determining the fluxes of particulate organic carbon (POC) between land and sea systems, providing vital ecological services (e.g., Bauer et al., 2013; Canuel and Hardison, 2016). POC in estuaries displays diverse reactivity with a wide range of ages, from modern to over 30,000 ^{14}C years depending on source, composition, and storage time and closely connected to many of the issues related to climate changes, and regional and global carbon cycles (e.g., Marwick et al., 2015; Tao et al., 2015). Under the modern condition, natural POC delivery systems from land to sea have been disturbed by human activities such as river damming (e.g., Bauer et al., 2013; Maavara et al., 2017). In South Korea, 228 estuaries have been perturbed by dam constructions for agricultural and flood control purposes (e.g., Lee et al., 2011). Such river impoundments have impacted estuary ecosystem such as phytoplankton community, food web structure, mollusk growth, richness of submerged macrophytes (Hong et al., 2007; Lee et al., 2018). Recently, social interest in ecological restoration and environmental conservation in estuaries has increased, raising issues about removal or opening of estuary dams (Suh et al., 2004; D. Kim et al., 2016). Studies are required to determine the source, composition, and reactivity of riverine POC for assessing potential changes in water quality and ecosystems upon removal or opening of estuary dams. However, there have been no studies on source-age characterization of POC along a land-sea interface in Korean estuaries.

In this study, we investigated the spatial variability of concentrations, and stable and radiocarbon isotopes of POC along a salinity gradient in two contrasting Korean estuary systems, closed (Geum) and open (Seomjin) estuary systems. Our main objectives were to determine the source and age of POC, and thus to evaluate the effect of an estuary dam on POC transfer to the adjacent coastal zone.

The Geum River, which flows into the mid-eastern Yellow Sea, is the third largest river in South Korea, with a drainage area of 9914 km^2 and a length of 398 km (Water Resources Management Information System, WAMIS). Its mean annual discharge in 2016 was 324 m^3/s, ranging from 102.4 m^3/s in February to 841.1 m^3/s in July (Water Environment Information System, WEIS). The Geum estuary is closed by a dam built in 1990. In contrast, the Seomjin River is an open estuary without a dam. The drainage basin area is 4914 km^2, and its main stem length is 222 km (WAMIS). Its mean annual discharge was 55.5 m^3/s in 2016, with a minimum value in August (21.9 m^3/s) and a maximum value in October (140.5 m^3/s) (WEIS).

Surface water samples were collected between the land (i.e., the last gauging station of each river) and sea end-member sites in the Geum and Seomjin estuaries in August 2016 (Fig. 1). All sample collections were conducted at or near high tide conditions. Surface water was collected directly into a high-density polyethylene carboy through Tygon tubing using an aspirator system. About 0.1–2 L of collected water was filtered through a pre-combusted (450 °C, 5 h) and pre-weighed 0.45-μm glass fiber filter (Macherey-Nagel, Dueren, Germany).

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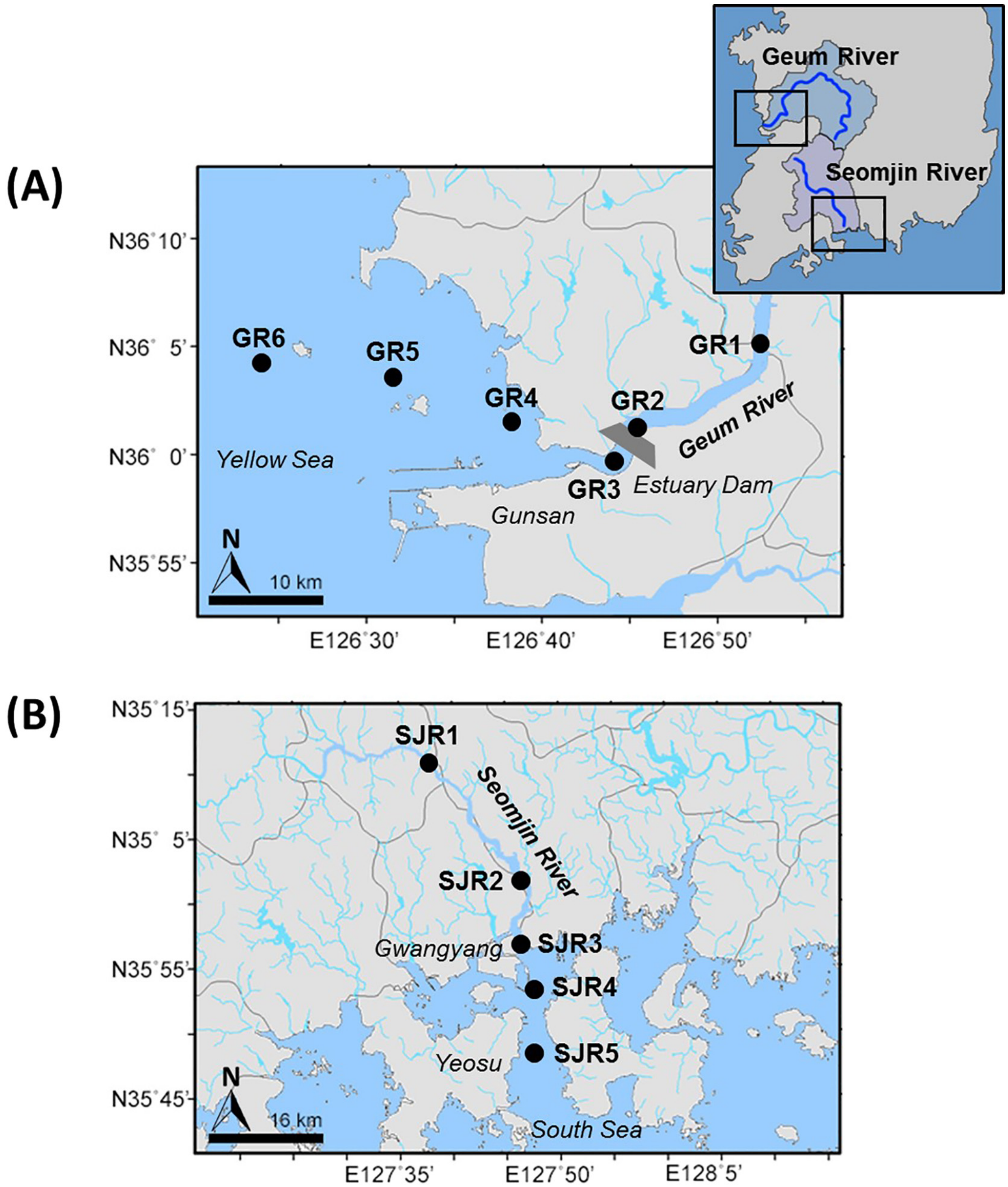

Fig. 1. Map showing the sampling locations in (A) the Geum estuary and (B) the Seomjin estuary.

Water parameters (temperature and salinity) were measured in-situ using a Hydrolab DS5 multi-parameter water quality sonde (OTT Hydromet, Kempten, Germany) during the sampling campaign. The filters were freeze-dried and weighed to calculate concentration of total suspended matter (TSM). To determine concentrations and stable isotopes of POC, inorganic carbon was removed using 12 M HCl under the fume hood. The POC and particulate nitrogen (PN) concentrations, and the stable isotope ratios of POC ($\delta^{13}C_{POC}$) were analyzed using an elemental analyzer combined with isotope ratio mass spectrometry (Delta V, Thermo Fisher Scientific, Bremen, Germany or Isoprime, GV Instrument, Manchester, UK). The $\delta^{13}C$ value was expressed in δ-notation relative to Vienna Pee-Dee Belemnite (VPDB). Radiocarbon analyses were conducted at the National Ocean Science Accelerator Mass Spectrometry Facility of the Woods Hole Oceanographic Institution (NOSAMS) in the USA and Alfred Wegener Institut (AWI) in Germany following standard routines. The radiocarbon data of POC were presented in *delta* notation ($\Delta^{14}C$, ‰).

In the Geum estuary, the average surface water temperature was 30.9 ± 1.1 °C, slightly decreasing toward the offshore direction (Fig. 2A). In contrast, salinity was drastically different before and after the dam, ranging from 0‰ to 34‰ (Fig. 2B). The TSM concentration was much higher just after the dam, with a value of 95.9 mg/L (Fig. 2C). The POC concentration showed a decreasing trend from the river to the sea site, varying between 0.19 mgC/L and 12.70 mgC/L (Fig. 3A). The POC/PN ratio ranged 8.1 to 16.9. $\delta^{13}C_{POC}$ and $\Delta^{14}C_{POC}$ were in the range of −23.9 to −19.4‰ and −98.2 to −48.1‰, respectively, with no distinctive difference before and after the dam (Fig. 3C–D). The data of temperature, salinity, POC concentration, POC/PN ratio and $\delta^{13}C_{POC}$ in GR1 were from Kang et al. (2019).

In the Seomjin estuary, the trend of surface water temperature was similar to that observed in the Geum estuary (Fig. 2A). However, salinity showed a more gradual increase from 0‰ to 35‰ (Fig. 2B). The TSM and POC concentrations did not show a trend, with average values of 23.10 ± 12.6 mg/L and 0.88 ± 0.1 mgC/L, respectively (Figs. 2C and 3A). The POC/PN ratio ranged from 9.2 to 26.6 (Fig. 3B). $\delta^{13}C_{POC}$ and $\Delta^{14}C_{POC}$ were lowest in the upper estuary, with values of −29.1‰

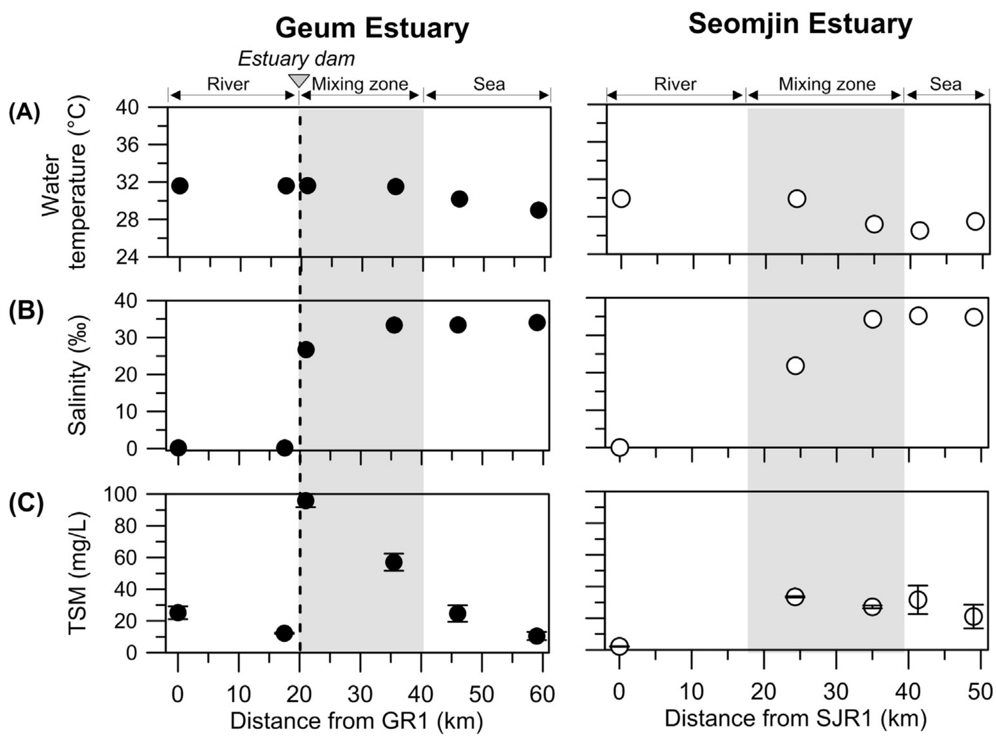

Fig. 2. Variation in water parameters: (A) water temperature (°C), (B) salinity (‰), and (C) TSM concentrations (mg/L). The data of water temperature, salinity in GR1 and SJR1 are from Kang et al. (2019).

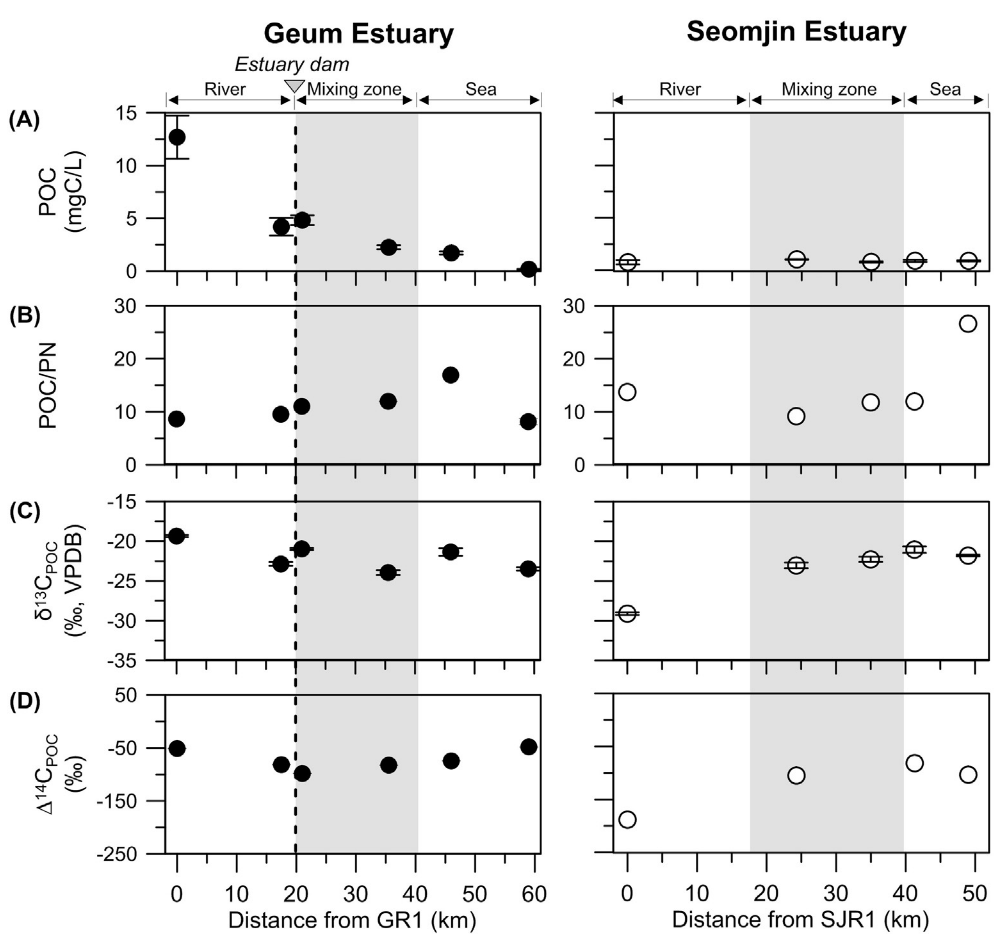

Fig. 3. Variation in (A) POC concentrations (mgC/L), (B) POC/PN ratio, (C) $\delta^{13}C_{POC}$ (‰), and (D) $\Delta^{14}C$ (‰) in the Geum and Seomjin estuaries.

and −188.3‰, respectively, and gradually increased toward the lower estuary, with values of −23.0 to −21.1‰ and −105.0 to −81.6‰ (Fig. 3C–D). The data of temperature, salinity, POC concentration, POC/PN ratio and $\delta^{13}C_{POC}$ in SJR1 were from Kang et al. (2019).

In a natural river system, POC concentration associated with soil erosion is commonly related to TSM concentration, which is strongly controlled by hydrodynamic processes (e.g., Wood, 1977; Guo et al., 2015; Wu et al., 2018). In the Seomjin estuary, POC concentration showed a positive correlation with TSM concentration ($R = 0.82$, $p = 0.09$), suggesting that input of soil-derived OC is an important contribution to the total POC pool. However, in an impounded river system, a dam transforms the natural hydrological conditions and material cycles (e.g., Baxter, 1977; Wu et al., 2007). In the Geum estuary, the dam alters the natural hydrodynamic processes and thus impacts POC concentration, leading to a weaker relationship with TSM concentration ($R = 0.60$, $p = 0.20$).

The POC%, calculated as POC/TSM × 100, has a negative relationship with TSM concentration in many natural estuary systems due to the dilution of POC by land-derived minerals (e.g., Meybeck, 1982; Ludwig and Probst, 1996; Guo et al., 2015). Such a negative correlation was observed between POC% and TSM in the open Seomjin estuary ($R = –0.67$, $p = 0.22$) but not in the closed Geum estuary ($R = 0.09$, $p = 0.87$). Notably, two samples (GR1 and GR2) from the reservoir of the Geum estuary had higher POC% but lower TSM concentrations, similar to the sample (SJR1) from the last gauging station of the Seomjin River, suggesting different sources of POC between Geum and Somjin estuaries, i.e., phytoplankton- vs. terrestrial-derived POC.

The POC/PN ratio in the Geum estuary was on average 9.1 ± 0.6 in the reservoir but slightly higher after the dam, with a value of 12.0 ± 3.7 (see Fig. 3B). Phytoplankton-derived organic matter has a POC/PN ratio lower than 10, but terrestrial-derived organic matter has a POC/PN ratio above 12 (e.g., Wu et al., 2007; Szczepańska et al., 2012). Hence, it appears that phytoplankton production in the reservoir of the Geum estuary contributed to the POC pool than after the dam. In contrast, in the Seomjin estuary, the POC/PN ratio was slightly higher at the SJR1 site (the last gauging station site) than at the other estuary sites but not the SJR5 site (see Figs. 1B and 3B). This suggests that the terrestrial-derived POC contribution was higher at the SJR1 site than at other estuary sites but not the SJR5 site.

In the Geum estuary, the $\delta^{13}C_{POC}$ values were in the range of −22.0 ± 1.7‰, without the gradual increasing trend that was observed in the Seomjin estuary (see Fig. 3C). The increasing $\delta^{13}C_{POC}$ trend was consistent with previous studies conducted in the Seomjin estuary (Kim et al., 2019) and other natural estuary systems (Wu et al., 2013; Guo et al., 2015). The low $\delta^{13}C_{POC}$ value of −29.1‰ in the most upstream site of the Seomjin estuary is comparable with the signatures of terrestrial C_3 plants, which use the Calvin pathway of carbon fixation with $\delta^{13}C$ values of −32 to −24‰ (e.g., Peterson and Fry, 1987; Meyers, 1997; Marwick et al., 2015). Indeed, the common C_3 reed (*Phragmites australis*) is dominant in the upstream part of the Seomjin estuary, with low $\delta^{13}C_{POC}$ values of −27 to −29‰ (Min and Je, 2002; Choi et al., 2005). On the other hand, the $\delta^{13}C_{POC}$ values of phytoplankton range from −23.5‰ to −18.9‰ in the Seomjin estuary (Kim et al., 2019) and from −24‰ to −16‰ in other natural estuary and marine systems (e.g., Lamb et al., 2006; McMahon et al., 2013; Guo et al., 2015). Hence, the increase of $\delta^{13}C_{POC}$ together with low POC/PN values toward the lower estuary sites in the Seomjin estuary indicate an increasing contribution of aquatic-derived POC to the total POC pool. Notably, seagrasses (*Zostera japonica* and *Zostera marina*) inhabiting the lower Seomjin estuary have high $\delta^{13}C_{POC}$ values of −8.6 to −10.1‰ (Kim et al., 2010; J.H. Kim et al., 2016; Kim et al., 2019) and high POC/PN values of 14.5 to 30.2 (J.H. Kim et al., 2016). Hence, the higher $\delta^{13}C$ value of −21.8‰ with a higher POC/PN ratio (26.6) at the lowest site of the Seomjin estuary (SJR5) appears to be due to a larger contribution of intertidal seagrasses (*Zostera japonica* and *Zostera marina*) to the total POC pool at this site. In contrast to the upstream site (SJR1) of the Seomjin estuary, the $\delta^{13}C_{POC}$ value in the reservoir of the Geum estuary (GR1 and GR2) was higher, with an average value of −21.1 ± 2.5‰, which was indistinguishable from that (on average −22.4 ± 1.5‰) after the estuary dam (see Fig. 3C). Freshwater phytoplankton has a wide range of $\delta^{13}C_{POC}$ values (−39 to −6‰) depending on CO_2 concentration and dissolved inorganic carbon isotopes (e.g., Meyers, 1997; Vuorio et al., 2006). Hence, $\delta^{13}C_{POC}$ values in the reservoir of the Geum estuary seem to be associated with increased phytoplankton production.

Evidence supporting the $\delta^{13}C_{POC}$ signatures discussed above can be found in the $\Delta^{14}C$ characteristics of POC, which have been commonly used for constraining the source and age of POC (e.g., Raymond and Bauer, 2001; Li et al., 2014; Marwick et al., 2015; Tao et al., 2015; Wu et al., 2018). In general, the $\Delta^{14}C$ of POC (i.e., $\Delta^{14}C_{POC}$) in rivers, estuaries, and coasts has a wide range of values from −1000‰ to +200‰ depending on mean time elapsed since biosynthesis and the integrated effect of transport or deposition of POC in the watershed (e.g., Raymond and Bauer, 2001; Galy et al., 2015; Marwick et al., 2015; Tao et al., 2018). The $\Delta^{14}C_{POC}$ in the Seomjin estuary (−119.6 ± 47.0‰) was lower than that of the Geum estuary (−66.4 ± 21.6‰ and −75.8 ± 20.9‰ before and after the dam, respectively). However, the $\Delta^{14}C_{POC}$ values from both Geum and Seomjin estuaries were higher than the global median signature of −230‰ in the estuaries (Marwick et al., 2015). Furthermore relatively younger POC was exported from the Geum and Seomjin estuaries in comparison to those from Chinese rivers that flow into the Yellow Sea, such as the Changjiang River (−436 to −103‰; Wang et al., 2012; Wu et al., 2018) and the Yellow River (−635 to −243‰; Tao et al., 2018; Wang et al., 2012) (see Fig. 4A). The $\Delta^{14}C_{POC}$ values of world rivers are generally lower when TSM increases but POC% decreases (see Fig. 4A and B), indicating that riverine POC is a mixture of organic-rich, fresher components such as litter or surface soil (higher POC% and $\Delta^{14}C_{POC}$ values but lower TSM concentration) and older, fossil components from bed rocks (lower POC% and $\Delta^{14}C_{POC}$ values but higher TSM concentration) (Marwick et al., 2015). In the Geum estuary, however, $\Delta^{14}C_{POC}$ showed a negative relationship with TSM ($R = –0.77$, $p = 0.07$) and no clear relationship with POC% ($R = 0.03$, $p = 0.96$), showing no clear mixing trend of two end-members, as shown in other world rivers. High POC% with high $\Delta^{14}C_{POC}$ were reported in Three Gorges Dam reservoirs in the Changjiang River due to fresher POC derived from in-situ phytoplankton production (Wu et al., 2018). Hence, the high $\Delta^{14}C_{POC}$ value associated with high POC% and $\delta^{13}C_{POC}$ values in the reservoir of the Geum estuary is similarly indicative of a larger contribution of fresher POC to the total POC pool due to in-situ phytoplankton production (see Fig. 5A and B). The $\delta^{13}C_{POC}$ value of −29.1‰ associated with $\Delta^{14}C_{POC}$ value of −188.3‰ at the SJR1 site of the Seomjin estuary is also indicative of a larger contribution of fresher POC to the total POC pool, but due to terrestrial, allochthones inputs, in contrast to the reservoir sites of the Geum estuary (see Fig. 5A and B). The lower sites of the Seomjin estuary (SJR2 to SJR5) showed lower values of POC% (on average 3.3 ± 0.7) but higher values of $\Delta^{14}C_{POC}$ (on average −96.7 ± 13.1‰) compared to those of the upstream site (SJR1), with a $\delta^{13}C_{POC}$ value of average −22.0 ± 0.8‰ (see Fig. 5A and B). This indicates that a contribution of fresher POC to the total POC pool was larger at the lower sites than at the upstream site of the Seomjin estuary, which is probably due to additional, fresher terrestrial contribution from the lower parts of the drainage basin.

The average modern POC content (modern C_{org} in wt%), calculated as POC% × F_m, where F_m represents the modern fraction in each sample (cf. Li et al., 2014), was 9.7 ± 12.5 wt% and 7.5 ± 11.8 wt% in the Geum and Seomjin estuaries, respectively (Fig. 5C). The reservoir samples of the Geum estuary (GR1 and GR2) had much higher modern POC contents, with the average value of 40.0 wt%, compared to those of the lower estuary (on average 4.1 wt% in GR3–6). The upper Seomjin estuary sample (SJR1) also showed a high value of the modern POC content (28.4 wt%), compared to those of the lower estuary (on average

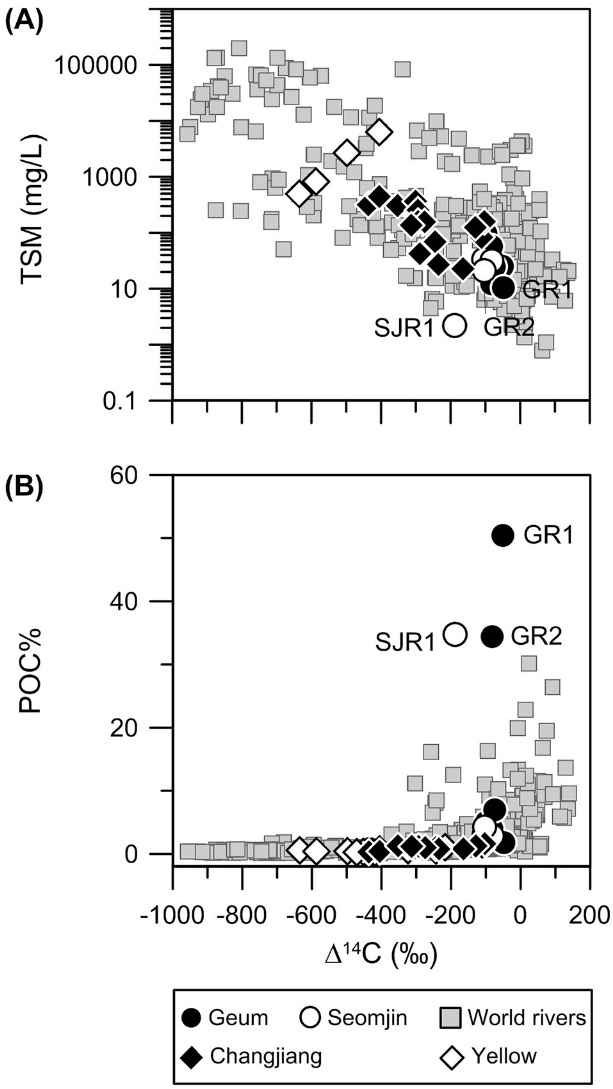

Fig. 4. Scatter plots of Δ^{14}C (‰) with (A) TSM (mg/L) and (B) POC%. The data from the Changjiang River, the Yellow River, and other world rivers are from Marwick et al. (2015), Xue et al. (2017), and Wu et al. (2018).

2.3 wt% in SJR2–5). The relative proportions of fossil and recently-fixed modern (terrestrial, riverine/estuarine, and marine) POC were also estimated using a simple binary mixing model as follows (cf. Yoon et al., 2016):

$$f_{\mathrm{fossil}} = (X_{\mathrm{sample}} - X_{\mathrm{modern}})/(X_{\mathrm{fossil}} - X_{\mathrm{modern}}) \times 100\% \quad (1)$$

where X_{sample} is the measured $\Delta^{14}C_{POC}$ value of the sample, and X_{fossil} and X_{modern} are the fossil ($\Delta^{14}C_{fossil}$) and modern ($\Delta^{14}C_{modern}$) end-member values of $\Delta^{14}C_{POC}$, respectively. We assumed $\Delta^{14}C_{fossil}$ as −1000‰ and $\Delta^{14}C_{modern}$ as 50‰ (cf. Wang et al., 2012). Our calculations showed that about 87–90% of modern POC existed in the reservoirs of the Geum estuary and about 86–91% of modern POC was present after the dam. In the Seomjin estuary, modern POC was 77–87%, with the highest proportion of 87% in the lower estuary (SJR4).

Accordingly, our results showed no distinct difference in δ^{13}C and Δ^{14}C before and after the dam of the Geum estuary, while the Seomjin estuary without a dam showed gradual changes. The modern POC contribution was higher (87–90%) in the Geum estuary reservoir than that (77%) in the upper Seomjin estuary. Our study thus highlights that an estuary dam can alter the source and age of POC, which can be exported to the lower estuary when the water gate is open and thus

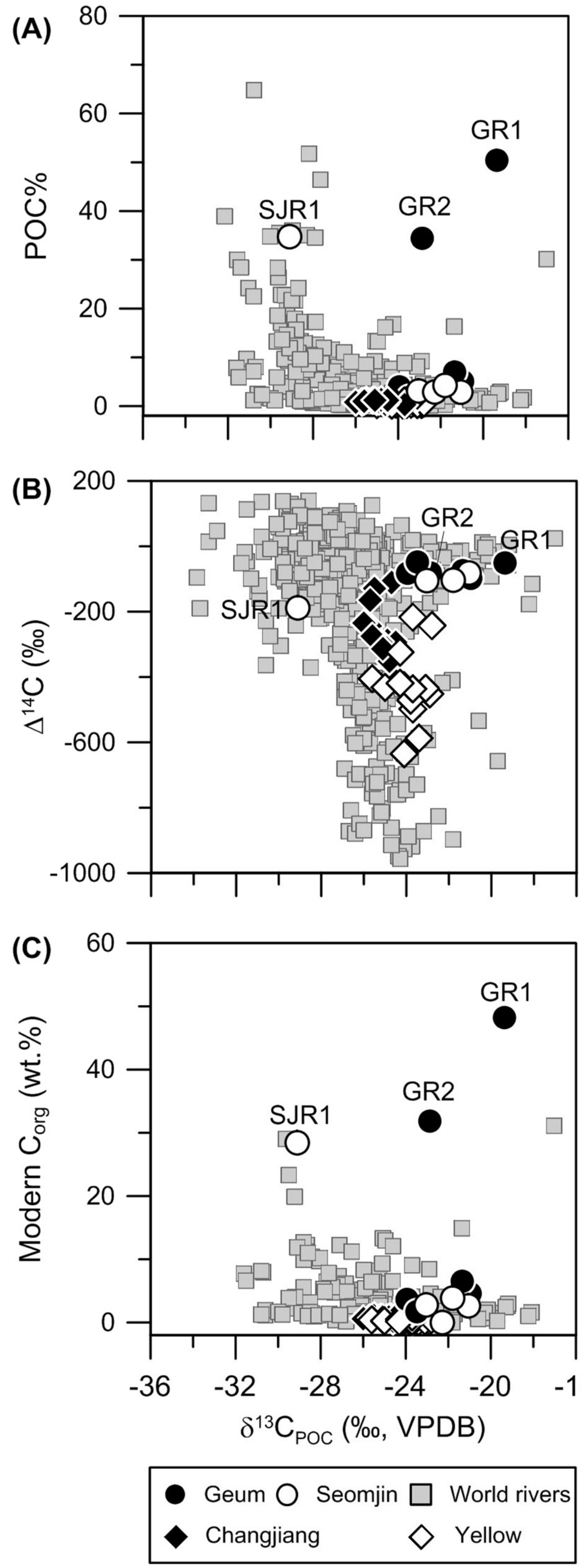

Fig. 5. Scatter plots of (A) POC%, (B) Δ^{14}C (‰), and (C) modern C_{org} (%) with $\delta^{13}C_{POC}$ (‰). The data from the Changjiang River, the Yellow River, and other world rivers were from Marwick et al. (2015), Xue et al. (2017), and Wu et al. (2018).

impact the adjacent coastal ecosystem.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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