Seasonal fluxes and source variation of organic carbon transported by two major Chinese Rivers: The Yellow River and Changjiang (Yangtze) River

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[1]   A one-year study was carried out to investigate the seasonal fluxes and source variation of organic carbon transported by two major Chinese rivers, the Yellow River and Changjiang. In 2009, the Yellow River and Changjiang transported 3.20×10^{10} g and 1.58×10^{12} g DOC and 3.89×10^{11} g and 1.52×10^{12} g POC, respectively. The dominant input of the terrestrial organic matter occurred during the high discharge period from June to July for the Yellow River and from June to August for Changjiang, accounting for 36–44% of the DOC and 72–86% of the POC transported by the two rivers in 2009. The Yellow River transported much higher concentrations of inorganic carbon than organic carbon, while a reverse trend was found in the Changjiang, indicating the different sources of carbon discharged by the two rivers. Using radiocarbon and stable carbon isotope measurements, we identified the different sources and seasonal variations of organic carbon transported by the Yellow River and Changjiang. The Yellow River carried old POC with radiocarbon ages ranging from 4000 to 8000 years, while POC transported by Changjiang had a relatively younger ^{14}C age ranging from 800 to 1060 years. The ^{14}C ages of DOC were relatively younger (305–1570 years) and showed less variation between the two rivers. The seasonal variations found in ^{14}C ages of DOC and POC indicate that a large fraction of recent-fixed labile organic carbon was transported by the two rivers in the spring and summer months. The different sources and seasonal variations in both fluxes and sources of organic carbon transported by the Yellow River and Changjiang could have an important influence on the biogeochemical cycle and ecosystems in the estuaries and adjacent coastal waters of the East China Sea.

1. Introduction

[2]   World rivers play important roles in the global carbon cycle by linking the land and ocean systems, which are the two largest carbon reservoirs on earth [*Dagg et al.*, 2004; *Bianchi and Allison*, 2009]. On a global scale, approximately 900 Tg (Tg = 1×10^{12} g) of carbon from various terrestrial sources including plants, soils and weathering rocks was transported by rivers to the oceans annually [see *McKee*, 2003, and references therein]. Among the total transported carbon, about 500 Tg is terrestrial organic carbon consisting 60% of dissolved organic carbon (DOC) and 40% of particulate organic carbon (POC) [*Spitzy and Ittekkot*, 1991; *Meybeck*, 1993; *Hedges et al.*, 1997]. It is estimated that 40% of the river-transported carbon is carried by the ten largest rivers in the world [*Milliman and Meade*, 1983; *Meade*, 1996; *Dagg et al.*, 2004]. Therefore, the large rivers, such as the Amazon, Mississippi and Changjiang (Yangtze), could play dominant roles not only for the regional climate and ecosystem, but also for the biogeochemical processes and carbon budget and cycle in the river-dominated marginal seas [*Meybeck*, 1982; *Degens et al.*, 1991; *McKee*, 2003; *Amon and Meon*, 2004; *Bianchi et al.*, 2007].

[3]   The sources and fate of terrestrial organic carbon transported by rivers are largely affected by the river drainage basins. For carbonate mineral-rich basins, high alkalinity and dissolved inorganic carbon (DIC) could be carried by the rivers such as observed for the Yellow River [*Chen et al.*, 2005; *Zhang and Zhang*, 2007] and Mississippi River [*Raymond and Cole*, 2003]. Organic matter transported by

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rivers usually show a very refractory and detritus feature consisting of highly decomposed organic matter from soils, vegetation and weathered materials [*Hedges*, 1992; *Dixon et al.*, 1994; *Lal*, 2003]. Riverine refractory organic matter is thus believed to play a less important role in the biogeochemistry of the estuaries and coastal oceans [*Lobbes et al.*, 2000; *Amon et al.*, 2003].

[4] Using radiocarbon measurement, *Raymond and Bauer* [2001a] have shown that rivers could export aged terrestrial organic matter to the ocean and the ^{14}C ages of POC varied significantly from 316 to 4,763 years (before present) in the rivers they sampled. The old ages of organic matter suggest the highly degraded and refractory nature of the terrestrial organic matter transported by rivers. For DOC, it appears that their ^{14}C ages are much younger than their counterpart POC in many world rivers such as the Amazon [*Raymond and Bauer*, 2001b] and some Arctic rivers including the Yukon, Mackenzie, Yenisey and Ob' Rivers [*Benner et al.*, 2004; *Guo and Macdonald*, 2006; *Raymond et al.*, 2007]. A recent study by *Holmes et al.* [2008] has shown that DOC transported by some Alaskan rivers could be remarkably labile during the spring flood period when the majority of annual DOC flux occurs. Their study suggests that the riverine inputs of labile DOC to the Arctic Ocean may have a much larger influence on coastal ocean biogeochemistry than previously thought. This certainly challenges our knowledge of the role of terrestrially derived DOC on carbon, microbial, and food-web dynamics in the coastal oceans. Since the drainage basin varies river by river, our knowledge of the temporal and spatial variations of carbon sources transported by the world's large rivers, however, is very limited, especially as different river drainage basins related to the regional continental environment and climate change [*McKee*, 2003].

[5] In this paper, we report the results from a one-year study carried out in 2009 to investigate the seasonal fluxes and source variations of organic carbon transported by the two major rivers in China, the Yellow River and Changjiang River. This work was part of a five-year joint national research project to study the carbon budget and cycle in China's marginal seas (http://973oceancarbon.xmu.edu.cn). The Changjiang and Yellow Rivers, together, are the major links between China's continent and the East China Sea (ECS), one of the largest continental marginal seas in the world, and thus play important roles in the carbon cycle and biogeochemical processes in ECS. In our study, we determined and compared the monthly and annual fluxes of DOC and POC carried by the two rivers in 2009. We also applied stable and natural radioactive carbon isotope (^{13}C and ^{14}C) measurements to identify and compare, for the first time, the sources and seasonal variations of terrestrial organic carbon transported by the Yellow River and Changjiang.

2. Materials and Methods

2.1. Study Site

[6] Changjiang, with its 6,300 km drainage length, is the largest river in China and the third longest river in the world [*Milliman and Meade*, 1983]. It originates from the glaciers on the eastern part of the Tibetan Plateau and drains one-fifth of China's continental area of more than 1.94×10^6 km^2 before emptying into the East China Sea (Figure 1). The low reach of Changjiang River passes through a temperate climate region of the southeast China where the terrestrial vegetation coverage is relatively abundant along both sides of the river. The Yellow River is the second largest river in China and the seventh longest in the world at 5,464 km in length. The Yellow River originates in the Bayanhar Mountain region in the far west of China and drains a basin area of 752,443 km^2 and discharges into the western Bohai Sea (Figure 1). The drainage basin of the Yellow River is more complex, encompassing a broad range of geological tectonic features including the oldest metamorphic rocks to modern fluvial-lacustrine sediments, carbonates and clastic rocks from the Paleozoic to Mesozoic age, and Quaternary loess deposits mainly in the middle reach of the river [*Zhang et al.*, 1995]. The Loess Plateau, the major source of the suspended particles to the Yellow River, is one of the largest and thickest loess deposits in the world [*Chen et al.*, 2005]. As one important feature, the Yellow River has been recognized as being one of the highest sediment load rivers on earth, at about 1×10^9 t/y [*Milliman and Syvitski*, 1992]. The Changjiang and Yellow Rivers together provide not only the major (80%) freshwater input to the ECS, but also a huge amount of terrestrial organic matter into the ECS, and thus play a dominant role in the carbon cycle and ecosystems in the estuaries and adjacent coastal waters.

2.2. Sampling

[7] Monthly samples were collected from the low reaches of both the Changjiang and Yellow Rivers in 2009. We selected Datong Hydrographic Station of Changjiang and Lijin Hydrographic Station of the Yellow River as our sampling sites (Figure 1). Datong Hydrographic Station is located about 500 km upstream from the river mouth and is the last monitoring station in the low reach of Changjiang before it enters the ECS. Lijin Hydrographic Station is the last monitoring station in the low reach of the Yellow River and is located about 80 km from the river mouth. The flow rates and concentrations of total suspended matter (TSM) as routine monitoring parameters are measured and recorded daily at both stations. Monthly average flow rates and TSM concentrations were calculated based on the daily measurements and provided by the two hydrographic stations. For our study, water samples were collected at both stations in the middle of each month in 2009. For the Yellow River, since the water depth of the main channel was relatively shallow (~1.5 m) during most months in recent years, duplicate water samples were collected each month from the middle depth in the central mainstream of the river. For Changjiang, the average water depth in the main channel of the sampling station was about 10 m and water samples were collected at three depths (subsurface, middle and deep) in the central mainstream of the river. After collection, the three depth waters were mixed in 2:1:1 volume to make a represent sample for the whole water column based on the TSM sampling procedure of the Datong Hydrographic Station. Duplicate samples were taken for DOC and POC measurements. All water samples were filtered through precombusted glass fiber filters (Whatman, 0.7 μM, 47 mm). Suspended particles retained on filters (usually several liters were filtered) were kept frozen for elemental (C and N) and C-isotopic (^{13}C and ^{14}C) analyses of POC. Water (0.5 L) for DOC and DO^{14}C measurements was acidified with 50% H$_3$PO$_4$ to pH 2 and kept frozen until analysis. All glassware
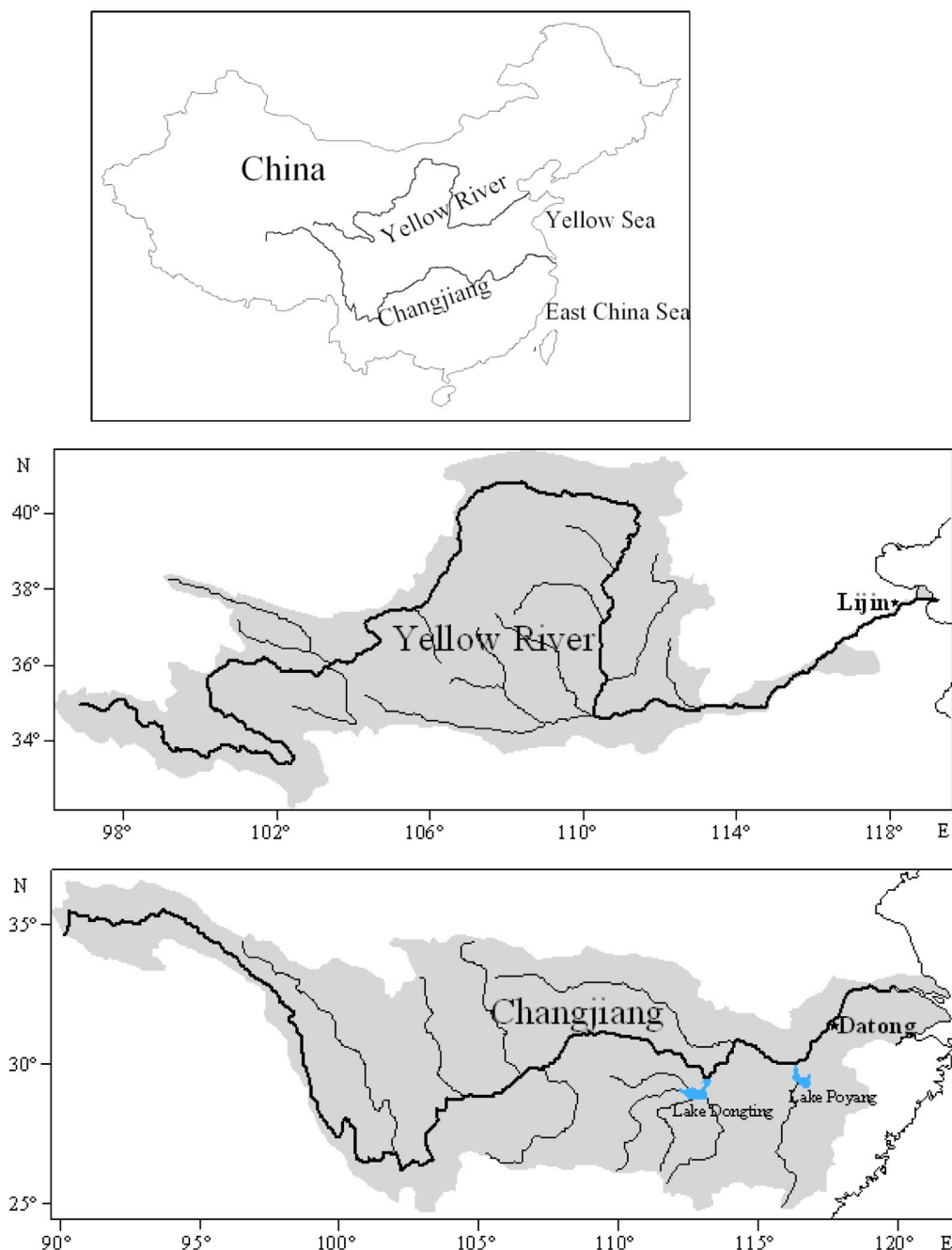
Figure 1. Map showing the Yellow River and Changjiang drainage basins and the locations of Lijin Hydrographic Station (low reach of Yellow River) and Datong Hydrographic Station (low reach of Changjiang) where samples were collected.

used for water sample collection, processing and storage were washed with 10% HCl acid and Milli-Q water and pre-combusted at 550°C for 6 h based on the established procedures [*Wang et al.*, 2004].

2.3. Measurement of DOC, POC, PIC and PN

[8] DOC concentrations were analyzed by the high temperature combustion (HTC) method using a Shimadzu TOC-V Total Organic Carbon Analyzer. DOC standard was prepared using potassium hydrogen phthalate (KHP) and UV-oxidized Milli-Q water. Both low carbon water and deep seawater (from D. Hansell's lab at the University of Miami) were used for instrument blank check and quality control during sample analysis [*Sharp et al.*, 2002]. Total blanks associated with DOC analyses were about 10 μM, which was, in general, <8% of the DOC concentrations of our river water samples. Analytic precision on triplicate injections was <3%.

[9] River suspended particles were analyzed for POC, particulate inorganic carbon (PIC) and particulate total nitrogen (PN) using a Perkin-Elmer 2400 CHNS Analyzer. Samples of suspended particles were analyzed separately for total carbon content and POC before and after acidification

Table 1. Monthly Flow Rate, TSM, Dissolved Organic Carbon (DOC) and Particulate Carbon (POC, PIC) and Nitrogen (PN) Concentrations Measured in the Low Reach of Yellow River and Changjiang River in 2009[a]

Month	Flow Rate (1000l/s)	TSM (kg/1000l)	DOC (μM)	POC (% dry wt.)	PIC (% dry wt.)	PN (% dry wt.)	C/N (mole)
			Yellow River				
January	196	0.495	174 ± 13	0.57 ± 0.05	1.83 ± 0.13	0.17 ± 0.03	4.1
February	115	1.05	170 ± 14	0.40 ± 0.04	1.55 ± 0.10	0.10 ± 0.02	4.6
March	176	1.24	190 ± 15	0.38 ± 0.06	1.56 ± 0.08	0.12 ± 0.02	3.8
April	154	0.825	176 ± 12	0.41 ± 0.04	1.69 ± 0.13	0.11 ± 0.04	4.7
May	216	1.08	171 ± 10	0.45 ± 0.07	2.07 ± 0.14	0.12 ± 0.03	4.6
June	982	10.60	151 ± 11	0.79 ± 0.08	2.90 ± 0.12	0.30 ± 0.03	3.0
July	944	6.30	231 ± 18	0.70 ± 0.06	2.96 ± 0.15	0.21 ± 0.02	5.9
August	380	1.17	280 ± 16	0.37 ± 0.03	2.26 ± 0.11	0.14 ± 0.03	4.5
September	509	2.00	225 ± 13	0.40 ± 0.04	1.83 ± 0.13	0.18 ± 0.05	3.8
October	572	2.69	212 ± 12	0.44 ± 0.05	1.94 ± 0.09	0.17 ± 0.04	4.4
November	466	1.95	218 ± 11	0.39 ± 0.03	1.65 ± 0.12	0.16 ± 0.03	4.0
December	328	0.866	226 ± 12	0.41 ± 0.06	1.91 ± 0.16	0.16 ± 0.04	4.1
			Changjiang River				
January	11600	0.083	180 ± 15	1.01 ± 0.10	0.31 ± 0.03	0.14 ± 0.02	8.6
February	11800	0.060	170 ± 14	0.96 ± 0.08	0.29 ± 0.02	0.12 ± 0.02	9.7
March	23600	0.079	165 ± 12	1.25 ± 0.09	0.36 ± 0.04	0.16 ± 0.04	9.1
April	23000	0.117	151 ± 11	1.12 ± 0.08	0.30 ± 0.04	0.12 ± 0.03	10.9
May	32300	0.077	175 ± 12	1.30 ± 0.10	0.24 ± 0.03	0.15 ± 0.03	10.1
June	35800	0.139	185 ± 14	1.43 ± 0.09	0.26 ± 0.04	0.14 ± 0.03	12.0
July	40000	0.159	228 ± 17	1.65 ± 0.13	0.28 ± 0.03	0.20 ± 0.02	9.6
August	42400	0.267	151 ± 10	1.35 ± 0.11	0.22 ± 0.04	0.24 ± 0.04	9.2
September	32800	0.234	137 ± 11	1.18 ± 0.10	0.30 ± 0.05	0.22 ± 0.04	8.9
October	17100	0.125	165 ± 10	1.27 ± 0.08	0.21 ± 0.02	0.21 ± 0.03	10.2
November	14000	0.188	167 ± 14	1.04 ± 0.08	0.24 ± 0.03	0.18 ± 0.03	9.4
December	12000	0.077	158 ± 10	0.98 ± 0.07	0.32 ± 0.04	0.18 ± 0.02	8.9

[a]The monthly flow rate and TSM concentrations were calculated based on the daily measurements of the month. Concentrations of DOC, POC, PIC and PN were average values of duplicate sample measurements.

(10% HCl). PIC content was calculated as the difference by subtracting POC from the total carbon content of each sample. C/N mole ratio was calculated based on the POC and PN measurements. The analytic precision based on replicated analysis was ±4% for TOC, PIC and ±5% for PN.

2.4. Carbon Isotopic Measurement

[10] Stable carbon (δ^{13}C) and natural radiocarbon (Δ^{14}C) abundances were measured for both DOC and POC for samples collected in four months (January, April, July and October) to determine and compare the seasonal changes of the sources and ages of organic matter transported by the two rivers. Analysis of both Δ^{14}C and δ^{13}C of the samples was performed at the National Ocean Science Accelerator Mass Spectrometry (NOSAMS) facilities at Woods Hole Oceanographic Institution (WHOI). Briefly, for isotopic measurement of DOC, filtered and acidified river water (usually 200–300 ml for each sample) were first UV-oxidized using a modified low-blank ultraviolet oxidation and vacuum line system built at NOSAMS based on *Beaupre et al.* [2007]. The system was then blank tested using standard materials to ensure satisfaction of both low blank and effective UV oxidation of water samples. The generated CO_2 from UV-oxidation of water samples was purified, collected and measured on the vacuum line and split for δ^{13}C and Δ^{14}C measurements. For POC isotopic measurement, samples were first acidified with 10% HCl to remove inorganic carbon and then dried at 50°C. The dried POC samples were combusted at NOSAMS and the generated CO_2 from combustion was collected and measured on the vacuum line and split for δ^{13}C and Δ^{14}C measurements. CO_2 gas for Δ^{14}C

measurement was reduced for graphite and Δ^{14}C was measured by AMS following established procedures at NOSAMS [*McNichol et al.*, 1995]. Values of δ^{13}C are reported in ‰ relative to the PDB standard and values of Δ^{14}C are reported as the fraction modern based on modern reference material used and conventional radiocarbon ages (year before present) were calculated based on *Stuiver and Polach* [1977].

3. Results and Discussion

3.1. Concentrations of DOC, POC and PIC

[11] Based on the measurements at the two hydrographic stations, the average monthly flow rates and concentrations of TSM ranged from 115 to 982 m³/s and 0.495 to 10.60 kg/m³ (dry wt.) for the Yellow River and 11,600 to 40,000 m³/s and 0.060 to 0.267 kg/m³ (dry wt) for Changjiang in 2009 (Table 1 and Figure 2). The average monthly flow rates of Changjiang were 40 to 100 times higher than for the Yellow River during 2009. The sediment load measured as TSM, however, was 8 to 40 times higher in the Yellow River than the TSM in Changjiang, indicating the highly turbid nature of the Yellow River. The high concentration of TSM transported by the Yellow River was largely due to the contribution from the Quaternary loess deposits along the middle reaches of the Yellow River [*Milliman and Syvitski*, 1992; *Zhang et al.*, 1990; *Chen et al.*, 2005]. As seen in Figure 2, the apparent high discharge period during June and July which accounted for 38% of the total annual fresh water flux of the Yellow River in 2009 was, in fact, a man-controlled event. Since the Xiaolangdi Dam was built in the middle reach of the Yellow River in 2001, the flow
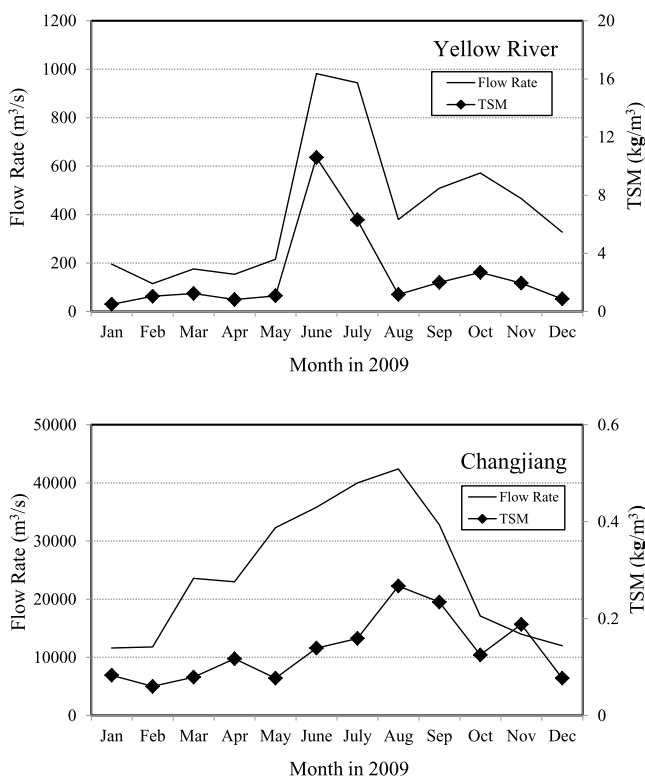
Figure 2. Monthly mean flow rate (m³/s) and TSM concentrations (kg/m³) measured at the Lijin Hydrographic Station and Datong Hydrographic Station for the Yellow River and Changjiang in 2009.

rate in the low reach of the river has been well controlled for flood events and also for large demand of water usage. The flow rate in the low reach of the Yellow River has been kept at a low level of 100–200 m³/s during most months of the year (data from Lijin Hydrographic Station) to avoid dryness in the low reach of the river and more importantly, to protect the ecosystems in the Yellow River Estuary and surrounding wetland. In late June and early July before the rainy season each year, the dam gates are scheduled to open and let the high flow flood and clean the low reach river bed as suspended particles deposit during the low flow period. As a result, very high TSM concentrations were measured during this short high flood period and that accounted for 56% of the total annual sediment flux (Figure 2). In comparison, the average monthly flow rate of Changjiang in 2009 was less affected by the Three Gorges Dam and was comparable to the previous years (data from Datong Hydrographic Station). However, studies have shown that even the flow rate of Changjiang at its low reach was less affected in the past years [*Wang et al.*, 2008]; the sediment discharge to the ECS by Changjiang has decreased significantly, up to 40% since the construction of the Three Gorges Dam [*Yang et al.*, 2006; *Wang et al.*, 2008].

[12] Concentrations of DOC, POC, PIC and PN measured for the Yellow River and Changjiang are summarized in Table 1. Monthly average concentrations of DOC are comparable and show less seasonal variations for the two rivers, ranging from 151 to 280 μM for the Yellow River and 137 to 228 μM for the Changjiang River. Concentrations of POC and PIC ranged from 0.37 to 0.79% and 1.55–2.90% (dry

wt.) for the Yellow River, and 0.96–1.65% and 0.21–0.36% for Changjiang, respectively. Concentrations of PN show similar values for the two rivers ranging from 0.10 to 0.30% for the Yellow River and 0.12–0.24% for the Changjiang. The calculated C/N ratios for the particles transported by the two rivers, however, show distinct differences. Particles in the Yellow River had much lower C/N ratios (3.0–5.9) with lower POC% than the particles carried by Changjiang (C/N = 8.6–10.9).

[13] The POC/DOC ratio has been reported for some rivers. Based on the differences of the drainage basins of the rivers, the POC/DOC export ratio ranged from 0.2 to 9.0 [*Degens et al.*, 1991]. *McKee* [2003] summarized that the latest studies converge a global river average POC/DOC ratio of ∼1.0, suggesting that a mass balance exists between river-transported POC and DOC. The overall mean POC/DOC ratio we determined for Changjiang is about 1.0, which is in good agreement with the global average value and is also consistent with the values reported by *Wu et al.* [2007], who measured that the POC/DOC ratio ranged from 0.81 to 1.1 in Changjiang in a 5-year study from 2003 to 2008. For the Yellow River, however, the average POC/DOC ratio reached 12, an order of magnitude higher than the values of Changjiang and other major world rivers such as the Amazon (0.68), Zaire (0.27) and Mississippi (0.23) [*Dagg et al.* 2004]. A low POC/DOC ratio could indicate that labile POC are biodegraded and released into DOC pool more easily, or dissolved more easily as affected by adsorption/desorption and chemical dissolution processes [*Keil et al.*, 1997; *Hedges et al.*, 2001]. On the other hand, the very high POC/DOC ratio found for the Yellow River may suggest that this riverine POC could be extremely refractory and bound tightly to mineral particles. These POC were less affected by biodegradation and dissolution processes, thus could have longer residence time during the transport in the river. As discussed in the later section, this explanation is also well demonstrated by our isotopic results.

[14] For TSM transported by the two rivers, another significant difference is that the PIC contents are several times higher for the Yellow River than for the Changjiang. These differences indicate that the particles transported by the two rivers are likely from very different sources as the two rivers flow through different drainage basins. For the Yellow River, highly decomposed loess deposits and carbonate minerals in the middle reach were likely the major sources contributing significantly to the TSM [*Zhang et al.*, 1990, 1992; *Chen et al.*, 2005]. *Zhang et al.* [2007] reported that in the Yellow River, particles with sizes of <32 μm, mainly clay minerals, accounted for >95% of TSM. These highly decomposed clay minerals had low content of organic carbon and C/N ratio. *Chen et al.* [2005] summarized 42 years (1958–2000) water chemistry records in the Yellow River and reported that Ca^{2+} and HCO_3^- were the dominant ion pair in the water and the concentrations of these ions in the Yellow River were the highest among the large rivers such as Amazon, Lena and Changjiang they compared, suggesting that significant contributions from carbonate minerals to the Yellow River occurred. For Changjiang River in comparison, a large fraction of TSM is derived from terrestrial plant detritus, which contributes to a relatively high organic C content and C/N ratios [*Wu et al.*, 2007; *Zhang et al.*, 2007]. Using lignin and stable carbon isotope analyses, *Yu*
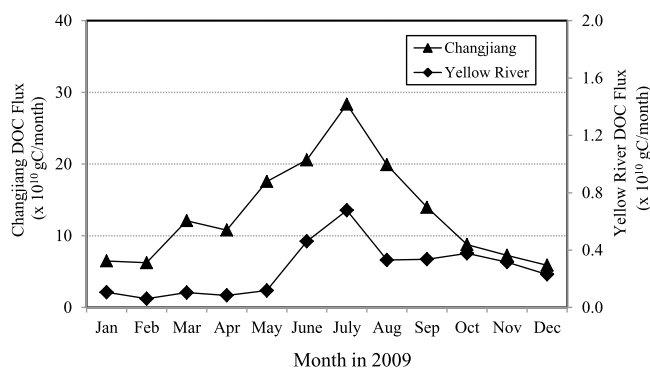
Figure 3. Calculated monthly fluxes of DOC in the Yellow River and Changjiang in 2009.

et al. [2011] recently reported that both the quantity and quality of particulate matter transported by the Changjiang River showed significant variation in different reaches of the river. In the low reach of the river, terrestrial plants appeared to be the major source of POM to the river. This is well expected since the low reach of Changjiang River flows through a temperate climate region in the southeast of China, terrestrial vegetation coverage along the river banks is more abundant and evergreen than the region of low reach of the Yellow River. These source differences are also well demonstrated by our carbon isotope data and are discussed further in a later section.
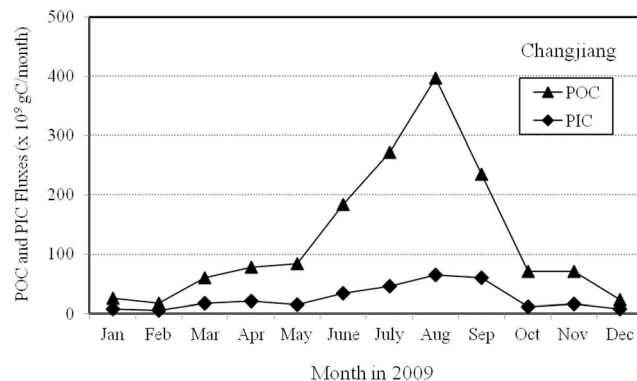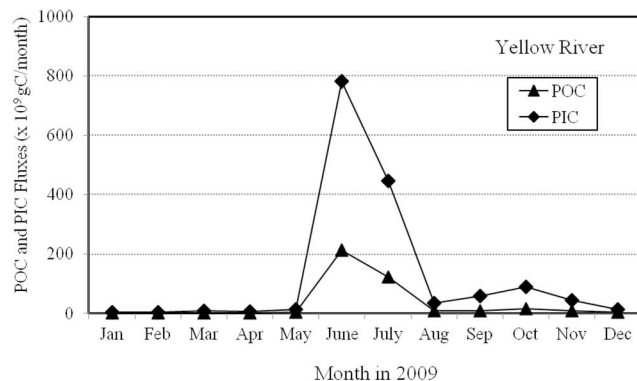

Figure 4. Calculated monthly fluxes of POC and PIC in the Yellow River and Changjiang in 2009.
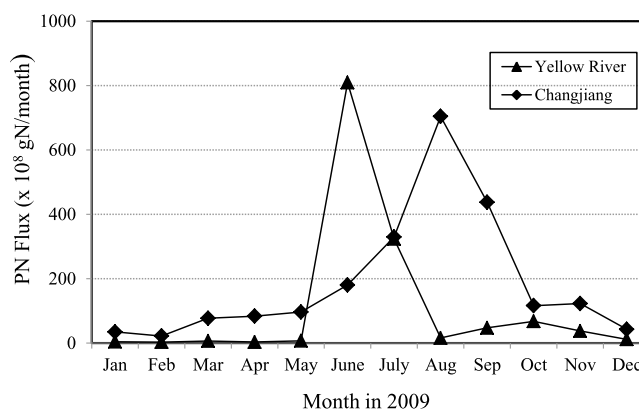

Figure 5. Calculated monthly fluxes of PN in the Yellow River and Changjiang in 2009.

3.2. Fluxes of DOC, POC and PIC

[15] Based on the monthly mean flow rates and TSM concentrations and the measured average concentrations of DOC, POC, PIC and PN, we calculated the monthly fluxes of DOC, POC, PIC and PN for the Yellow River and Changjiang in 2009. As plotted in Figure 3, the monthly fluxes of DOC in Changjiang ranged from 5.89×10^{10} to 28.34×10^{10} gC, which was 40–100 times higher than the monthly DOC fluxes in the Yellow River during 2009. The peak flux of DOC appeared from the period of June to July for the Yellow River and from June to August for the Changjiang, corresponding well with the monthly flow rate of the two rivers. The peak flux accounted for 36% of the annual DOC flux in the Yellow River and 44% of the annual DOC flux in Changjiang in 2009. For POC and PIC, the high fluxes also occurred from the period of June to July in the Yellow River and from June to September for the Changjiang, which accounted for 86% and 81%, and 72% and 67% of the annual POC and PIC fluxes carried by the two rivers in 2009, respectively (Figure 4). Due to the higher concentrations of PIC than POC of the transported particles, the higher fluxes of PIC than POC were observed in every month, especially during the high flux period for the Yellow River. In contrast, POC fluxes were more dominant in every month in the Changjiang, consistent with the different source supplies of suspended particles in the two rivers as discussed above. Figure 5 shows the monthly fluxes of PN in the two rivers. PN fluxes in Changjiang ranged from 2.20×10^9 to 70.42×10^9 gN/month and were higher than the values in the Yellow River in most months except during the flood period. The high flux of PN during the flood period in the Yellow River was mainly due to the very high load of TSM in that month (Figure 2).

[16] Based on the monthly fluxes, we calculated the annual fluxes of DOC, POC, PIC and PN for the two rivers. In 2009, the Yellow River transported 3.20×10^{10} g DOC, 3.89×10^{11} g POC, 1.51×10^{12} g PIC and 1.34×10^{11} g PN into the Baohai Sea, and Changjiang delivered 1.58×10^{12} g DOC, 1.52×10^{12} g POC, 3.06×10^{11} g PIC and 2.25×10^{11} g PN into the East China Sea. The total terrestrial OC (POC + DOC) transported by the Yellow River and Changjiang were 4.21×10^{11} gC/yr and 3.1×10^{12} gC/yr in 2009, respectively. It should be emphasized that the uncertainties of our flux calculations came from two sources. The first was due to the

analytical errors of duplicate samples which can be quantified as shown in Table 1. For example, the sum of analytical errors of monthly duplicate samples could contribute up to ±8%, ±10% and ±12% uncertainties to the calculated annual DOC, POC and PIC fluxes. The major uncertainties for our estimated annual fluxes, however, could be due to the limited samples collected during the year. This could be particularly true for POC and PIC. Unlike the flux of DOC which was mainly controlled by water discharge rate, fluxes of POC and PIC could be largely affected by the concentration of suspended particles which is subject to the influences of gravitational setting, hydrodynamic lift and drag forces during river transport [*Battin et al.*, 2008]. Unfortunately, we are not able to quantitatively determine these uncertainties associated with our flux estimations. For the Yellow River, since the flow rate was relatively small and water depth was shallow (<1.5 m) during most months in 2009, flux variation with water depth was not concerned. For Changjiang River, however, since the flow rate was much higher and the water depth in the center of the mainstream was deep (∼10 m), POC and PIC fluxes could vary with depth. Although our monthly samples were mixed water from three depth collections, it may not fully represent the suspended particle concentrations in the whole water column. A depth integrated fluxes for POC and PIC should be conducted in the future for comparison.

[17] To compare our flux estimations with the literature values for the two rivers, it appeared that the OC fluxes we determined for 2009 are significantly lower than the historical values but comparable with the flux values of the recent years reported for the two rivers. For the Yellow River, the data on fluxes of organic carbon is rather limited. *Zhang et al.* [1992] first reported POC and DOC fluxes of 6.1×10^{12} gC/yr and 2.0×10^{11} gC/yr for 1987, calculated based on samples collected in the two months of May (dry season) and September (wet season). These values are 6 times higher for DOC and 15 times higher for POC than the fluxes we determined in 2009. Similarly, based on the two-season data collected in the low reach of the Yellow River, *Cauwet and Mackenzie* [1993] estimated the POC flux of 4.5×10^{12} gC/yr and DOC flux of 6.0×10^{10} gC/yr for 1983. The POC flux was also 11 times higher than our value but the DOC flux was in the same order of magnitude as our value. More recently, *Zhang* [2004] studied POC and DOC fluxes in the Yellow River during low (August) and high flood (September) months. The values reported are slightly higher but comparable with our average low and high flood months. These flux differences reported are largely due to the changes of the flow rates of the Yellow River in these different years. Also, the calculated annual fluxes of *Zhang et al.* [1992] and *Cauwet and Mackenzie* [1993] were only based on two months of data, which could introduce significant uncertainties to their values. Since the flow rate has been decreased significantly and remained relatively constant in the low reach of the Yellow River in the last 6 years (data from Lijin Hydrographic Station), our one-year monthly study and calculations thus more accurately reflect the carbon fluxes of the Yellow River in the recent years. For the Changjiang River, there are number of studies estimated the carbon fluxes. In a recent paper, *Wu et al.* [2007] compared these annual fluxes and reported that the annual fluxes of both DOC and POC have decreased significantly from the early years. For DOC, the annual flux decreased from 2 to 3×10^{12} gC/yr to

0.9×10^{12} gC/yr, and POC flux decreased from an average of 5×10^{12} gC/yr to 2×10^{12} gC/yr. from 1960 to 1980 to 1997, about 60% decrease for both DOC and POC fluxes in Changjiang. The 1997 annual fluxes of DOC and POC in Changjiang River determined by *Wu et al.* [2007] are actually comparable with the annual fluxes of DOC and POC we measured in 2009, indicating that the fluxes of OC transported by Changjiang have remained relatively constant in the recent years mainly due to the controlled flow rate and reduced TSM load by the Three Gorges Dam [*Yang et al.*, 2006; *Yu et al.*, 2011].

[18] Compared with the organic carbon flux in the major world rivers [*Dagg et al.*, 2004], the modern fluxes of POC and DOC in Changjiang still remain in the top ten largest rivers and POC flux in the Yellow River is also comparable to several large rivers in the world such as the Mississippi (USA), Lena (Russia) and Niger (Africa). For example, the annual fluxes of DOC (1.58×10^{12} g/yr) and POC (1.52×10^{12} g/yr) transported by Changjiang in 2009 are 51% and 163% of the values carried by the Mississippi River (DOC = 3.1×10^{12} g/yr, POC = 0.93×10^{12} g/yr) as recently reported by *Bianchi et al.* [2007]. Together, the total riverine organic C (POC + DOC) transported by the Changjiang and Yellow Rivers was 3.5×10^{12} gC/yr in 2009. This represents only 0.7% of the total organic carbon transported annually to the ocean by the world rivers (5.0×10^{14} gC/yr), small fraction but could be significant to the regional carbon and biogeochemical cycle in China's continents and the ECS. Comparing to the estimated primary production (1.44×10^{14} gC/yr) in the ECS [*Chen and Wang*, 1999], the total riverine organic C transported by the two rivers also accounts a small fraction (2.4%) of organic C produced in the ECS. Since less work has been done in the region, we know very little about the importance of terrestrial organic C, as well as what happens to this terrestrial organic C after entering the ECS. More studies need to be conducted in the future to investigate and compare the fate and biogeochemical processes of the riverine organic C transported by the two large rivers into the estuarine and coastal waters.

3.3. Source and Seasonal Variations of DOC and POC

[19] We measured $\delta^{13}C$ and $\Delta^{14}C$ for both DOC and POC for four selected months (January, April, July and October) to determine and compare the sources of DOC and POC and their seasonal changes of organic inputs as carried by the two rivers (Table 2). Values of $\delta^{13}C$ ranged from −25.6‰ to −32.1‰ and −28.8‰ to −32.1‰ for DOC and −23.4‰ to −25.6‰ and −23.1‰ to −24.7‰ for POC, in the Yellow River and Changjiang, respectively. DOC and POC transported by the two rivers had similar $\delta^{13}C$ mean values but DOC in both rivers was more depleted in $\delta^{13}C$ than POC. In their studies, *Wu et al.* [2007] and *Yu et al.* [2011] reported similar $\delta^{13}C$ values (−24.3‰ to −26.8‰) for POC collected in the mainstreams of the lower to upper reaches of Changjiang. These $\delta^{13}C$ values reflect the typical stable carbon isotope source signatures of terrestrial organic matter, similar to the values reported for the other large river systems such as the Amazon [*Hedges et al.*, 2000; *Raymond and Bauer*, 2001a, 2001b; *Townsend-Small et al.*, 2007], Mississippi [*Wang et al.*, 2004], and Yukon River and several other large arctic rivers [*Guo and Macdonald*, 2006; *Neff et al.*, 2006; *Raymond et al.*, 2007]. In comparison, the

Table 2. Carbon Isotope (δ^{13}C, Δ^{14}C) Measurements of DOC and POC Collected in the Low Reach of the Yellow River and Changjiang River[a]

	DOC				POC			
Sample Date	FM	Δ^{14}C (‰)	Age (year)	δ^{13}C (‰)	FM	Δ^{14}C (‰)	Age (year)	δ^{13}C (‰)
				Yellow River				
January 20	0.8755	−131	1070	−30.1	0.3676	−635	8040	−24.1
April 20	0.9323	−74	565	−28.9	0.4153	−587	7050	−23.4
July 20	0.9515	−55	400	−32.1	0.5993	−405	4110	−25.6
October 20	0.8840	−122	990	−25.6	0.5055	−498	5480	−23.7
Mean	0.9108	−96	756	−29.2	0.4719	−531	6170	−24.2
				Changjiang				
January 20	0.8224	−183	1570	−28.8	0.8954	−111	885	−24.7
April 20	0.8955	−110	885	−30.3				
July 20	0.9626	−44	305	−32.2	0.9034	−103	815	−23.1
October 20	0.9111	−95	745	−29.8	0.8767	−129	1060	−25.5
Mean	0.9070	−108	876	−30.3	0.8918	−114	920	−24.4

[a]Changjiang April 20 POC was lost during process.

values of Δ^{14}C measured for both DOC and POC showed distinct differences between the two rivers. As plotted in Figure 6, the ^{14}C ages of POC transported by the Yellow River are extremely old, ranging from 4,110 to 8,040 years, while the DOC are relatively younger with ^{14}C ages ranging from 400 to 1,070 years. In Contrast, the ^{14}C ages of DOC in Changjiang ranged from 305 to 1570 years, comparable to the values of the Yellow River DOC. However, the ^{14}C ages of POC in Changjiang (815–1060 years) are much younger than the POC ages of the Yellow River. These distinct ^{14}C age differences of DOC and POC found in the two rivers clearly indicate that the input sources of POC to the two rivers are different. The very old POC carried by the Yellow River suggests that these POC were not from the recent-fixed freshwater plankton and terrestrial plant materials. Rather, these POC were primarily derived from the highly decomposed soil, clay minerals and quaternary loess in the middle reach of the river [*Zhang et al.*, 1995], and perhaps weathering of old rocks and ancient kerogen. The unmatched ^{14}C ages of POC and DOC in the Yellow River also support our discussion earlier that those old POC were extremely refractory, bond tightly with the clay particles, and could not be easily released into the DOC pool from either chemical dissolution/desorption or biological processes during the river transport. In comparison, the relatively young ^{14}C age POC in Changjiang and DOC in both rivers suggest that these carbon pools contained a mixture of both old and a large fraction of recent-fixed modern terrestrial organic materials. This great difference of ^{14}C ages between DOC and POC has also been reported for other river systems. For example, *Raymond and Bauer* [2001a] measured Δ^{14}C of DOC and POC in the Amazon and Hudson Rivers and a few small rivers (York, Parker) in the Northeast of the United States. They found that the ^{14}C ages of DOC were all younger than POC in these rivers. DOC had a more modern ^{14}C age than POC (1,258 years BP) in the Amazon, and POC in the Hudson River was 3,000 years older than the riverine DOC (1,380 year BP). Young terrigenous DOC was also measured in several Arctic rivers such as the Yenisey, Mackenzie and Yukon rivers [*Amon and Meon*, 2004; *Benner et al.*, 2004; *Guo and Macdonald*, 2006; *Neff et al.*, 2006; *Raymond et al.*, 2007]. These studies suggest that the predominant sources of organic matter that contributed to the

young ages of riverine DOC were from the leaching and decomposition of recent-fixed carbon in plant litter and upper soil horizons. The ^{14}C age differences found between POC and DOC also support our discussion above suggesting that the two OC pools represent two different organic sources and they are isotopically disjointed. POC and DOC pools are regulated by chemical and biological processes at different
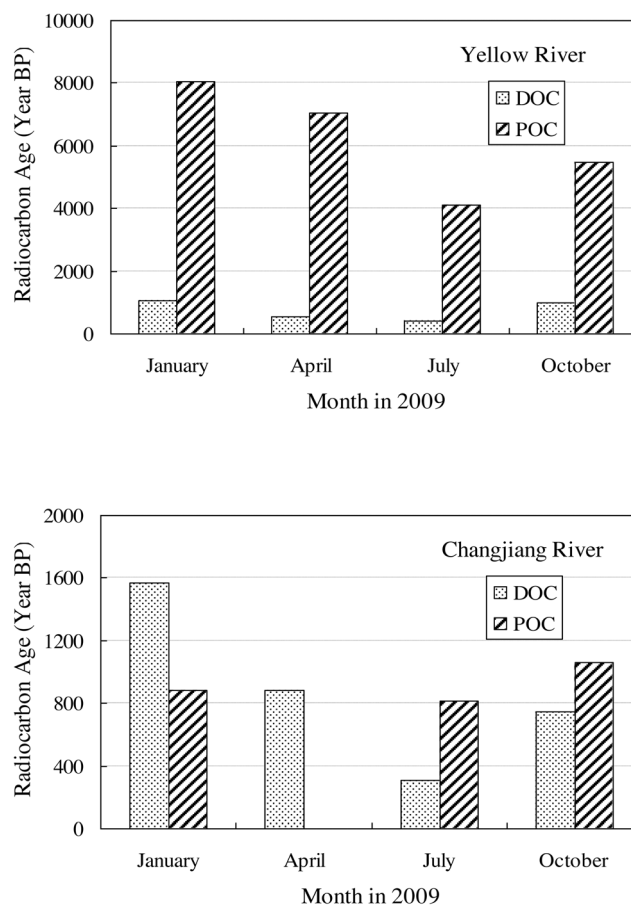

Figure 6. Plot of radiocarbon ages (year before present) for DOC and POC collected in January, April, July and October in the Yellow River and Changjiang in 2009.
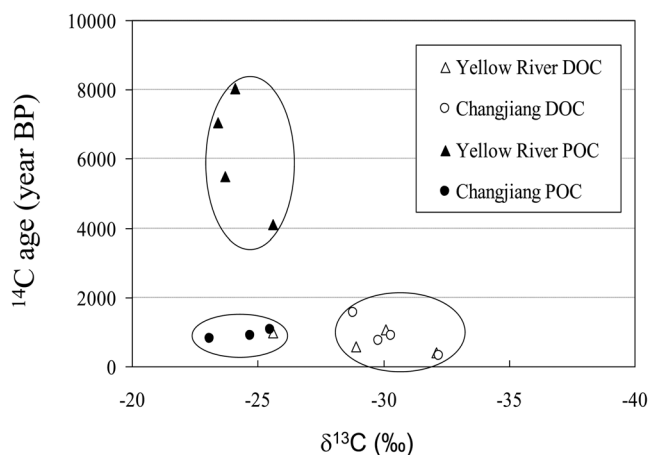
Figure 7. Plot of ^{14}C age versus δ^{13}C for DOC and POC collected from the Yellow River and Changjiang.

rates and timescales during their transport in the rivers. In Figure 7, we also plotted the correlation of δ^{13}C and ^{14}C ages for DOC and POC. It is quite clear that POC transported by the Yellow River and Changjiang can be separated into two groups with similar δ^{13}C values but different ^{14}C ages. DOC in the two rivers (with one exception) joined in one group with relatively young ^{14}C age and more depleted ^{13}C values, consistent with our discussion above on the source variations of organic matter transported by the two rivers.

[20] The sources of POC and DOC transported by the Yellow River and Chanjiang also showed strong seasonal variations (Figure 6). The ^{14}C ages of POC in the Yellow River decreased from winter (January) to the spring (April) and summer (July) and then increased again in the late fall. This observed decrease in the POC ages was likely due to the contribution of some recent-fixed organic carbon from freshwater plankton and terrestrial plants during the spring and summer months when the primary production is high. In late fall when the contribution of the modern ^{14}C decreased, the POC age then increased. In response, we see the same trend of seasonal changes for the ^{14}C ages of DOC in the Yellow River, indicating that some young organic carbon was added into the DOC pool as well in the spring and summer months. In comparison, the seasonal variation was more pronounced for DOC than POC in the Changjiang River in 2009. The ^{14}C ages of DOC decreased rapidly from winter to the spring and summer months and then increased again in the late fall. The POC ages, however, remained relatively constant with the season, suggesting that the sources of POC to Changjiang were primarily from the terrestrial plants and perhaps mixed with some old organic carbon from the upper soil deposits. In their series studies, *Bianchi et al.* [2004] and *Duan and Bianchi* [2006] investigated the seasonal variability in sources of DOC and POC transported in the low Mississippi and Pearl Rivers. They found that in situ phytoplankton production played an important role for the observed seasonal variations of DOC and POC. Based on the lignin and ^{13}C NMR analyses, they indicated that autochthonous production in the rivers could be more significant than previously thought which could affect on the age and lability of riverine organic matter entering the ocean. The more rapid seasonal changes of the ^{14}C ages of DOC than

POC found in the Yellow and Changjiang rivers in our study could also suggest that the microbial turnover rate of DOC was much faster than POC and the terrestrial DOC had much shorter resident time than POC during the transport processes in the rivers. Since the ^{14}C ages of DOC in the Yellow River showed less seasonal variation than DOC in Changjiang, it is also possible that relatively low light intensity or high shading effect in the turbid Yellow River water could limit the phytoplankton production in some degrees [*Duan and Bianchi*, 2006], resulting in less young aged DOC production during the spring and summer months.

[21] Since the recent-fixed plant organic matter has a young radiocarbon age (assume Δ^{14}C = +50‰) and is labile, we can estimate how much recent-fixed labile organic carbon was added to the POC and DOC pools in spring and summer months assuming that the winter (January) POC and DOC were relatively refractory (as measured aged Δ^{14}C values). Our calculations indicate that at least 7% and 34% recent-fixed C and 31% and 42% recent-fixed C had been added to the POC and DOC pools in April and July in the Yellow River; 31% and 60% recent-fixed C were contributed to the DOC pool in April and July in Changjiang. Although the transformation and fate of these labile and refractory terrestrial organic carbon transported by the Yellow River and Changjiang have not been well studied, our study suggests that the seasonal changes in organic sources and the contributions of labile and refractory POC and DOC by the two rivers could have an important influence not only on the biogeochemical and ecosystem processes in the estuaries and coastal waters, but also on the carbon cycle and budget in the ECS as well. The differences in organic source inputs and seasonal variations of POC and DOC transported by the Yellow River and Changjiang provide a perfect comparison to study and address this important yet still unanswered question.

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