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Spatial distribution of riverine DOC inputs to the ocean: an updated global synthesis

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On the basis of most up-to-date literature data, this study evaluated the influxes of dissolved organic carbon (DOC) to the ocean. Using the DOC concentrations in 118 world rivers and long-term average river discharges, we estimated the river influxes of DOC to the coastal seas as 0.21 Pg C yr^{-1} . This flux was reduced to 0.19 Pg C vr⁻¹ when we took into account DOC removal during its transport into the Arctic. When we further adopted an average removal rate of 10% for the rest of the river DOC input into the coastal ocean, we obtained an estimate of the global river DOC flux of 0.17 Pg C yr⁻¹, which is at the lower end of prior estimations. Considering the seasonal variation of the river end-member DOC concentration, our current estimate of the global river DOC discharge is subject to an uncertainty of ~30%.DOC fluxes into the ocean have significant spatial variations in terms of their continents of origin, recipient coastal seas, ocean basins and latitudinal zones. The highest DOC flux was from South America into the western ocean boundaries and eventually into the Atlantic Ocean. The most abundant riverine DOC discharge was in the low latitudinal zones with 38.0 and 90.0 Tg C yr $^{-1}$ in the 0–30 $^{\circ}$ N and 0–30 $^{\circ}$ S zones, respectively, the combination of which accounted for ${\sim}62\%$ of the global DOC input. On the basis of these updated fluxes, we estimated a global river mean DOC concentration of 5.29 mg L^{-1} .

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Introduction

Riverine organic carbon concentrations and their variations reflect terrestrial ecosystem changes, while their export into the ocean is an important constraint of the oceanic carbon budget and cycling. There have been many attempts throughout the past few decades to approach this riverine organic carbon flux $[1-7,8^{\circ},9,10^{\circ\circ},11]$ either via the fluvial concentrations or organic carbon losses from terrestrial ecosystems (Table 1).

Degens and Ittekkot [12] estimate the global dissolved organic carbon (DOC) input from rivers to the coastal seas as 0.11-0.25 Pg C yr⁻¹. A more recent assessment by Cauwet [9], taking into account data from the Arctic rivers collected in the 1990s, determines a DOC flux of 0.25 Pg C yr⁻¹. Cai [13] adopts a DOC flux of 0.25 Pg C yr⁻¹ primarily based on the data from Meybeck [4,5].

Another approach to estimate riverine DOC fluxes is based on the loss rate of soil organic carbon from the drainage basin. Ludwig *et al.* [7] note that the DOC flux is mainly a function of discharge, basin slope, and the carbon content of the soils in the drainage basin, and estimate an annual riverine DOC flux to the coastal oceans of 0.21 Pg C yr⁻¹. Along this line, Harrison *et al.* [10^{••}] predict that 0.17 Pg C yr⁻¹ is exported by rivers to the coastal zones as DOC. Aitkenhead and McDowell [8[•]], using annual fluxes of DOC from 164 watersheds, which were grouped into 15 biome types, discover a strong linear relationship between the mean annual fluxes of DOC and the mean soil C/N ratios of those biomes. Their linear model predicts a DOC flux to the coastal seas of 0.36 Pg C yr⁻¹, which is higher than other estimates.

Schlesinger and Melack [1] estimate the transport of total organic carbon (TOC) in the world's rivers as 0.37 Pg C yr⁻¹ by inventorying and extrapolating data on losses of carbon per unit volume of river discharge from 12 intermediate and large rivers. They also derive a slightly higher number, 0.41 Pg C yr⁻¹, from measurements of fluvial losses of organic carbon per unit area of land in various ecosystem types. If we assume an equal partitioning [3,6,13] between particulate organic carbon (POC) and DOC in TOC, the riverine DOC transport to the coastal seas would be around 0.19–0.21 Pg C yr⁻¹.

Taken together, the estimated range of riverine DOC fluxes into the coastal oceans is still quite large (Table 1). Major challenges in reliably estimating riverine DOC fluxes are related to a few issues. First of all, most of the above estimates are based on a similar data set from the world's major large rivers with other rivers occasionally included. The estimates also neglect the biogeochemical reactions within the estuaries and sometimes

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Riverine dissolved organic carbon (DOC) flux to the global coastal ocean.							

Reference	Flux (Pg C yr ⁻¹)	Remarks
Meybeck, 1981, 1982 [3,4]	0.22	Typology method based on the average DOC contents in various climatic zones
Mantoura and Woodward, 1983 [2]	0.78	Extrapolated based on the DOC flux of the Severn River catchment only, assuming that the global DOC export accounts for 1% of the world terrestrial primary production
Smith and Hollibaugh, 1993 [6]	0.20	Assigned a nominal value for the riverine DOC flux based on previous estimates
Ludwig et al., 1996 [7]	0.21	Based on an empirical relationship between the observed organic carbon
		fluxes and the climatic, biologic, and geomorphologic patterns characterizing the river basins
Aitkenhead and McDowell, 2000 [8*]	0.36	Based on DOC fluxes from 164 watersheds, which were grouped into 15 biome types, and established a relationship between DOC flux and soil C:N ratio on this biome basis
Cauwet, 2002 [9]	0.25	Based on the DOC fluxes from world large rivers and the data from the Arctic rivers collected in the 1990s
Harrison <i>et al.</i> , 2005 [10**]	0.17	Based on the Global Nutrient Export from Watersheds (NEWS) Model
Seitzinger et al., 2005 [11]	0.17	Based on the Global Nutrient Export from Watersheds (NEWS) Model
Cai, 2011 [13]	0.25	Adopted based on previous estimates
This study	0.17	Based on the DOC concentrations in 118 world rivers and long term average river discharges with consideration of estuarine DOC removal

those within the upper river depending on the location where the DOC concentrations were measured. Furthermore, these estimates do not take into account the dynamic seasonal changes in DOC concentrations in the river end-members when synthesizing at the global scale.

During the past few decades, there has been a significant accumulation of DOC data from river and estuarine systems all over the world, and it is thus timely to synthesize the most updated DOC fluxes into the ocean. It is also fortunate that we now have a very comprehensive river discharge data set $[14^{\bullet\bullet}, 15^{\bullet\bullet}]$, which will clearly improve our estimates. This paper thus seeks to adopt the most updated river discharge data and riverine DOC concentration data from the world's rivers in order to reexamine DOC fluxes. We emphasize the spatial distribution of riverine fluxes in terms of latitude, continent, ocean basin, and their immediate recipient — the coastal sea.

We will also attempt to adjust the net input fluxes into the coastal seas by considering the estuarine behavior of DOC given the recently recognized significant removal processes of DOC within the Arctic regime. Moreover, under global climate changes and the likelihood of intensified hydrological circulation, we have seen the redistribution of fresh water discharges into different ocean basins [15^{••}]. Therefore, we will also assess the potential changes of riverine DOC fluxes induced by such redistribution of fresh water discharge during the past few decades.

Coastal sea classification and data sources Coastal sea classification

It is clear that the coastal sea has a high concentration of boundary exchanges and processes, and represents the most dynamic region both in terms of physics and biogeochemistry (e.g. Robinson and Brink [16]; Liu *et al.* [17]). In this study, we adopted the classification of Robinson and Brink [16], whereby the world's coastal seas are grouped into four major categories: western and eastern ocean boundaries, polar, semi-enclosed seas/ islands and Australia (Figure 1). Adjustments have been made to assign the East China Sea as a western boundary current marginal sea and the entire South China Sea as a semi-enclosed marginal sea.

The western ocean boundaries are characterized by strong western boundary currents, relatively wider shelves, and abundant river discharges [18-20]. The eastern ocean boundaries are clearly marked by upwellings and narrow shelves [21]. Polar ocean coastal boundaries, notably the Arctic, are characterized by abundant river discharge, wider shelves, as well as strong seasonality [22]. The semi-enclosed seas are characterized by limited exchanges with open oceans and thus possess relatively longer water residence times [23], which allows for biogeochemical reactions to occur, and anthropogenic signals may also be accumulated. Although the yearly DOC discharged into the coastal ocean $(0.17-0.78 \text{ Pg C yr}^{-1})$ Table 1) is small compared to the total oceanic DOC pool (662 Pg C) [24], it represents important heterogeneous loadings and thus cannot be downplayed [9]. We therefore summarized the fluxes into these pan-regions for use in future studies considering the river-estuary-coastal ocean and open ocean interior as a carbon continuum. Given the fact that the coastal sea typically has a higher DOC production rate, the freshly produced DOC therein, when exported to the open ocean interior, may serve as an important carbon pump [25]. At the same time, DOC transport and transformation through the river-coastal sea-open ocean continuum remains the primary challenge in understanding global carbon cycling.





The world's major river systems and shelf/marginal seas (1. Arabian Sea; 2. Arafura Sea; 3. Baffin Bay; 4. Baltic Sea; 5. Banda Sea; 6. Barents Sea; 7. Bay of Bengal; 8. Bay of Biscay; 9. Beaufort Sea; 10. Bering Sea; 11. Black Sea; 12. Bohai Sea; 13. Caribbean Sea; 14. Celebes Sea; 15. Celtic Sea; 16. Chukchi Sea; 17. Coral Sea; 18. East China Sea; 19. East Siberian Sea; 20. Greenland Sea; 21. Gulf of Alaska; 22. Gulf of California; 23. Gulf of Guinea; 24. Gulf of Mexico; 25. Gulf of St Lawrence; 26. Hudson Bay; 27. Java Sea; 28. Kara Sea; 29. Labrador Sea; 30. Laptev Sea; 31. Mediterranean Sea; 32. North Sea; 33. Norwegian Sea; 34. Persian Gulf; 35. Red Sea; 36. Sea of Japan; 37. Sea of Okhotsk; 38. South China Sea; 39. Sulu Sea; 40. Yellow Sea).

Freshwater discharge

The total global freshwater discharge, excluding that from Antarctica and Greenland, is about 37 288 \pm 662 km³ yr⁻¹. This estimate is based on river discharges from 921 rivers, or 73% of the total global discharge, and the records of the river discharges were from 1948 to 2004 [14^{••},15^{••}].

We categorized the 925 rivers from Dai *et al.* [15^{••}] based upon their spatial distribution in terms of different continents, coastal ocean classifications, ocean basins, and latitudes. In each category, we estimated the subtotal river discharge and its proportion to the total discharge of the 925 rivers from Dai *et al.* [15^{••}], and then we upscaled the subtotal discharge of each category to a global scale. The details of the calculation are found in the caption to Figure 2.

We then estimated the respective annual discharge from Africa, Asia, Oceania, Europe, North America, and South America as 3596, 9838, 592, 2162, 6271 and 14 828 km³ yr⁻¹; and the total annual discharge to the western boundaries, eastern boundaries, polar boundaries, semi-enclosed seas/islands and Australia was estimated to be 19 152, 5348, 5735 and 7052 km³ yr⁻¹. Most of the river discharge to the western boundaries comes from the Amazon River which provides 5390 km³ yr⁻¹

[15^{••}]. Annual fresh water discharge into the four ocean basins, the Arctic, Atlantic, Indian and Pacific was 3658, 20 006, 4532 and 9092 km³ yr⁻¹. Finally, these river discharges were distributed between different latitudes in terms of their river mouth locations and were 8902 km³ yr⁻¹ at 0–30° N, 14 179 km³ yr⁻¹ at 0–30° S, 7826 km³ yr⁻¹ at 30–60° N, 1483 km³ yr⁻¹ at 30–60° S, and 4898 km³ yr⁻¹ at 60–90° N. Again, the tropical Amazon River was the source of the large discharge to the 0–30° S zone, even though the river plume turns largely into the 0–30° N zone.

DOC data sources and flux estimation

The DOC data used in this synthesis were from 118 rivers and were updated to the measurements in 2009 with many data collected in the past 40 years. Whenever possible and justifiable, more up to date data were adopted. For example, Richey *et al.* [26] report an average DOC concentration of 4.46 mg L⁻¹ in the Amazon River, while according to Coynel *et al.* [27], the average DOC concentration of the Amazon is 5.70 mg L⁻¹ (calculated from DOC flux and river discharge). Similarly, the average DOC concentration of the Mississippi River is 8.79 mg L⁻¹ in one previous study [28], but 4.2 mg L⁻¹ from Dubois *et al.* [29], the value which we adopted in this study.



Figure 2

Spatial distribution of total river discharge (blue), average DOC concentration (red), and DOC flux (green) in terms of their river mouth origins by continent (a), recipient shelf seas (b), ocean basins (c), and latitudinal zones (d). The calculation of the total river discharge was as follows, using the example of African rivers. First, we considered all of the African rivers with available discharge data listed in Dai *et al.* [15^{••}]. Then, we summed up the discharges of these African rivers (called the subtotal) and calculated the subtotal's proportion of the total discharge of the 925 rivers. Because the total discharge of the 925 rivers was less than the global total discharge, we then upscaled the subtotal discharge of African rivers to a global scale, assuming that the proportion of the total discharge of African rivers in global total discharge was equal to the proportion of the subtotal discharge of African rivers.

(a) AF: Africa; AS: Asia; OC: Oceania; EU: Europe; NA: North America; SA: South America. The discharge from the respective continent was based on 84 rivers with a discharge of 3596 km³ yr⁻¹ from Africa; on 167 rivers with a discharge of 9838 km³ yr⁻¹ from Asia; on 86 rivers with a discharge of 592 km³ yr⁻¹ from Oceania; on 187 rivers with a discharge of 2162 km³ yr⁻¹ from Europe; on 193 rivers with a discharge of 6271 km³ yr⁻¹ from North America; and on 206 rivers with a discharge of 14 828 km³ yr⁻¹ from South America. The average DOC concentration and DOC flux were estimated to be $8.03 \pm 1.89 \text{ mg L}^{-1}$ and $28.9 \pm 6.8 \text{ Tg C yr}^{-1}$ from Africa based on 11 rivers; $5.32 \pm 0.29 \text{ mg L}^{-1}$ and $52.4 \pm 2.8 \text{ Tg C yr}^{-1}$ from Asia based on 26 rivers; $4.76 \pm 1.60 \text{ mg L}^{-1}$ and $2.82 \pm 0.95 \text{ Tg C yr}^{-1}$ from Oceania based on two rivers; $7.71 \pm 0.64 \text{ mg L}^{-1}$ and $16.7 \pm 1.4 \text{ Tg C yr}^{-1}$ from Europe based on 25 rivers; $4.22 \pm 0.13 \text{ mg L}^{-1}$ and $26.5 \pm 0.8 \text{ Tg C yr}^{-1}$ from North America based on 49 rivers; and $5.47 \pm 1.85 \text{ mg L}^{-1}$ and $81.1 \pm 27.4 \text{ Tg C yr}^{-1}$ from South America based on five rivers.

(b) W: western ocean boundaries; E: eastern ocean boundaries; P: polar ocean boundaries; S: semi-enclosed seas, islands and Australia. See text for details on the classification of the shelf/marginal seas. The discharge into the respective shelf/marginal sea types was based on 322 rivers with a discharge of 19 152 km³ yr⁻¹ into the western ocean boundaries; on 158 rivers with a discharge of 5348 km³ yr⁻¹ into the eastern ocean boundaries; on 115 rivers with a discharge of 5735 km³ yr⁻¹ into the polar ocean boundaries; and on 326 rivers with a discharge of 7052 km³ yr⁻¹ into the semi-enclosed seas, islands and Australia. The average DOC concentration and DOC flux into the respective shelf/marginal sea types were estimated to be $5.02 \pm 0.55 \text{ mg L}^{-1}$ and $96.1 \pm 10.4 \text{ Tg C yr}^{-1}$ in western ocean boundaries based on 35 rivers; $5.97 \pm 1.00 \text{ mg L}^{-1}$ and $31.9 \pm 5.4 \text{ Tg C yr}^{-1}$ in eastern ocean boundaries based on 28 rivers; and $4.60 \pm 0.22 \text{ mg L}^{-1}$ and $32.5 \pm 1.5 \text{ Tg C yr}^{-1}$ in semi-enclosed seas, islands and Australia based on 38 rivers.

(c) The discharges into the Arctic, Atlantic, Indian, and Pacific Oceans were 3658, 20 006, 4532, 9092 km³ yr⁻¹, respectively (see Dai and Trenberth [14**], Table 4). The average DOC concentration and DOC flux into the respective ocean basin were estimated to be $8.36 \pm 0.72 \text{ mg L}^{-1}$ and $30.6 \pm 2.6 \text{ Tg C yr}^{-1}$ into the Arctic based on 12 rivers; $5.85 \pm 0.36 \text{ mg L}^{-1}$ and $117 \pm 7 \text{ Tg C yr}^{-1}$ into the Atlantic based on 70 rivers; $4.19 \pm 0.51 \text{ mg L}^{-1}$ and $19.0 \pm 2.3 \text{ Tg C yr}^{-1}$ into the Indian based on 11 rivers; and $3.36 \pm 0.19 \text{ mg L}^{-1}$ and $30.5 \pm 1.7 \text{ Tg C yr}^{-1}$ into the Pacific based on 25 rivers. (d) The discharge into the respective latitudinal zone was based on 203 rivers with a discharge of 8902 km³ yr^{-1} at 0-30° N; on 221 rivers with a discharge of 14 179 km³ yr^{-1} at 0-30° S; on 312 rivers with a discharge of 7826 km³ yr^{-1} at 30-60° N; on 84 rivers with a discharge of 1483 km³ yr^{-1} at 30-60° S; and on 103 rivers with a discharge of 4898 km³ yr^{-1} at 60-90° N. The average DOC concentration and DOC flux into the respective latitudinal zone were estimated to be $4.27 \pm 0.29 \text{ mg L}^{-1}$ and $38.0 \pm 2.6 \text{ Tg C yr}^{-1}$ at $0-30^\circ$ N based on 28 rivers; $6.35 \pm 1.43 \text{ mg L}^{-1}$ and $9.0.\pm 20.2 \text{ Tg C yr}^{-1}$ at $0-30^\circ$ S based on nine rivers; $3.87 \pm 0.12 \text{ mg L}^{-1}$ and $30.3 \pm 0.9 \text{ Tg C yr}^{-1}$ at $30-60^\circ$ N based on 63 rivers; $5.20 \pm 2.32 \text{ mg L}^{-1}$ and $7.71 \pm 3.44 \text{ Tg C yr}^{-1}$ at $30-60^\circ$ S based on three rivers; and $8.02 \pm 0.60 \text{ mg L}^{-1}$ and $39.3 \pm 3.0 \text{ Tg C yr}^{-1}$ at $6-90^\circ$ N based on 15 rivers. These 118 rivers with available DOC concentrations accounted for $\sim 48\%$ of the global total discharge, and covered $\sim 54\%$ of the global total drainage land areas. These data are summarized in supplementary material.

The DOC flux calculations showing the spatial distribution from different continents, latitudes, and/or into different coastal oceans or ocean basins were based on individual river DOC fluxes and their summation in each category. We first calculated the discharge weighted average DOC concentration based on the summation of individual river discharges and DOC fluxes of each category. Such average DOC concentrations were then multiplied by the total river discharge of each category (see 'Freshwater discharge' section) to estimate the DOC flux. Finally, global river DOC flux was obtained by adding up all of the fluxes in each category.

DOC influxes from rivers

Figure 2 presents the spatial distribution of DOC fluxes in terms of the continent from which the river mouth originates, the recipient coastal seas and ocean basins. Also presented are the DOC fluxes distributed in different latitudes. Such spatial distribution of DOC input is critically important for examining the terrestrial sources of organic carbon and their potential linkage with oceanic carbon cycling because any redistribution of these fluxes would impact on the reactivity of the oceanic DOC pool and potentially the microbial community in the ocean that is fueled by DOC.

Riverine DOC fluxes from different continents

As shown in Figure 2(a), the discharge weighted average DOC concentration in rivers originating from Africa was $8.03 \pm 1.89 \text{ mg L}^{-1}$ based on 11 river DOC concentrations. The value was $5.32 \pm 0.29 \text{ mg L}^{-1}$ in Asia, $4.76 \pm 1.60 \text{ mg L}^{-1}$ in Oceania, $7.71 \pm 0.64 \text{ mg L}^{-1}$ in Europe, $4.22 \pm 0.13 \text{ mg L}^{-1}$ in North America, and $5.47 \pm 1.85 \text{ mg L}^{-1}$ in South America. The high DOC concentration in African rivers may be associated with the vegetation they flow through and the overall low basin slope [27]. In Europe, the relatively high river DOC concentration might be related to the relatively strong impact of anthropogenic activities [30]. Correspondingly, the DOC fluxes were 28.9 ± 6.8 , 52.4 ± 2.8 , 2.82 ± 0.95 , $16.7 \pm 1.4, 26.5 \pm 0.8, \text{ and } 81.1 \pm 27.4 \text{ Tg C yr}^{-1}$, respectively, with the highest fluxes emanating from South America and Asia. The summation of the annual global riverine DOC flux was $208 \pm 28 \text{ Tg C yr}^{-1}$ or $0.21 \pm$ $0.03 \text{ Pg C yr}^{-1}$.

Riverine DOC fluxes into different shelf/marginal seas

The average DOC concentration of the rivers to the western boundaries continental margins was estimated to be $5.02 \pm 0.55 \text{ mg L}^{-1}$, $5.97 \pm 1.00 \text{ mg L}^{-1}$ to the eastern boundaries continental margins, $7.94 \pm 0.44 \text{ mg L}^{-1}$ to the polar boundaries, and $4.60 \pm 0.22 \text{ mg L}^{-1}$ to the semi-

enclosed seas (Figure 2(b)). Rivers flowing into the polar regions had the highest DOC concentrations. In decreasing order, the DOC fluxes were 96.1 ± 10.4 , 45.5 ± 2.5 , 32.5 ± 1.5 , and 31.9 ± 5.4 Tg C yr⁻¹ into the western boundaries, polar boundaries, semi-enclosed seas, and eastern boundaries. The total DOC flux was then estimated to be 206 ± 12 Tg C yr⁻¹ or 0.21 ± 0.01 Pg C yr⁻¹.

Riverine DOC fluxes in different ocean basins

The average DOC concentration of the rivers flowing into the Arctic, Atlantic, Indian and Pacific Oceans were 8.36 ± 0.72 , 5.85 ± 0.36 , 4.19 ± 0.51 , and $3.36 \pm 0.19 \text{ mg L}^{-1}$ and, as expected, rivers flowing into the polar regions generally had the highest DOC concentrations due to the high organic carbon content in the basin soil and permafrost [31]. In decreasing order, the DOC fluxes were 117 ± 7 , 30.6 ± 2.6 , 30.5 ± 1.7 , and $19.0 \pm 2.3 \text{ Tg C yr}^{-1}$ into the Atlantic, Arctic, Pacific, and Indian Oceans (Figure 2(c)). Total DOC flux was then estimated to be $197 \pm 8 \text{ Tg C yr}^{-1}$ or $0.20 \pm 0.01 \text{ Pg C yr}^{-1}$.

Riverine DOC fluxes in different latitudinal zones

The overall trend in terms of DOC concentration was as follows: the highest DOC occurred in high latitudes and lower DOC in lower latitudes excluding the $0-30^{\circ}$ S zone, where the Amazon River had a high DOC and dominated the zonal DOC concentration and flux (Figure 2(d)). The most abundant riverine DOC discharges occurred in the low latitudinal zones with 38.0 ± 2.6 and $90.0 \pm$ $20.2 \text{ Tg C yr}^{-1}$ in the 0–30° N and 0–30° S zones, the combination of which $(0.13 \pm 0.02 \text{ Pg C yr}^{-1})$ accounted for 62% of the global DOC input. This estimate is comparable to Huang *et al.*'s result of 0.136 Pg C yr⁻¹ ([32], this issue). In the 30–60° N and 30–60° S bands, the flux was 30.3 ± 0.9 and 7.71 ± 3.44 Tg C yr⁻¹. At the high latitudes of 60–90° N, DOC flux was $39.3 \pm 3.0 \text{ Tg C yr}^{-1}$. Note that riverine input was negligible at high latitude in the southern hemisphere. The extrapolated global DOC flux was 205 ± 21 Tg C yr⁻¹ or 0.21 ± 0.02 Pg C yr⁻¹.

Discussion

Global mean river DOC concentration and uncertainties associated with our flux estimates

No matter how we extrapolated, the DOC influx from rivers remained tight, ranging from 0.20 to 0.21 Pg C yr⁻¹. This reflected the fact that DOC flux is predominantly determined by river discharge. The global average river DOC concentration was thus estimated to be 5.29 ± 0.22 mg L⁻¹. This is slightly lower than the prior estimate of 5.75 mg L⁻¹ [4].

Many river DOC concentrations are subject to strong seasonality which, as mentioned above, represents one of the major uncertainties in estimating DOC fluxes. For example, the seasonal change in DOC concentrations in the Changjiang is $\sim 40\%$ (Lin J, MS thesis, Xiamen University, 2007). The Sepik River has a DOC seasonal

variability of ~13% [33], and the Mississippi River a seasonal DOC change of up to ~30% [29,34]. To obtain a conservative estimation, here we assigned a seasonal variation of 30% to the DOC river end-member at the global scale based on the references (Lin J, MS thesis, Xiamen University, 2007; and [29,33,34]), and, given an estimated uncertainty of river discharge as 2% [15^{••}], we determined an uncertainty of global river DOC discharge as ~30%. Note that such assigned seasonality of DOC river end-member concentration was based on a very limited data set from several large rivers, which should be better constrained.

Removal of DOC in the Arctic estuaries/shelves and elsewhere

Dittmar and Kattner [31] estimate that the total riverine DOC input into the Arctic Ocean is $18-26 \text{ Tg C yr}^{-1}$ which is similar to that of the Amazon, and they also suggest that there are practically no removal mechanisms in the estuaries and shelves. Raymond *et al.* [35] note a higher estimate of 25–36 Tg C yr⁻¹. In our study, we estimated that the total DOC flux discharged by rivers into the Arctic Ocean was 31 Tg C yr⁻¹ based on 12 large Arctic rivers.

Recent studies observe substantial removal of terrestrial DOC (tDOC) in the Arctic shelves [36^{••},37], and the first order tDOC decay constant, λ , is estimated to be $0.097 \pm 0.004 \text{ yr}^{-1} [36^{\bullet\bullet}] \text{ or } 0.06 \text{ yr}^{-1} [37] \text{ for the western}$ Arctic. An earlier study [38] also observes a significant portion of DOC in the colloidal fraction both at the river end-member and during estuarine mixing, and a nonconservative behavior both in the Ob and Yenisey Rivers and in the Kara Sea. The removal rate is close to 30% in the Ob River estuary [38]. On the basis of Δ^{14} C-DOC, Raymond et al. [35] reveal that Arctic rivers export a large amount of young and presumably semi-labile DOC to the Arctic Ocean. More recently, Letscher et al. [39**] report a much more rapid removal rate constant of 0.24 ± 0.07 yr⁻¹ in the eastern Arctic Ocean, and further estimate that the total tDOC input of 25 Tg C yr⁻¹ [35] into the Arctic Basin is reduced to 5.3–10.3 Tg C yr^{-1} before being transported to the North Atlantic with a residence time of several years. Alling *et al.* [40] report DOC removals of up to 10–20% in the Lena River estuary and the Laptev Sea with a surface water residence time of ~ 2 months, while such removals could be 30-50% along the East Siberian Sea shelf with freshwater residence times of several years.

Taken together, we reasoned that a conservative estimate of DOC removal in the Arctic estuaries would be around 20% and the total riverine DOC input into the Arctic Ocean, that is, 31 Tg C yr^{-1} , would be reduced to 24.5 Tg C yr⁻¹. Such a removal term accounts for approximately 3% of the global total DOC flux into the coastal seas. With this estimate, the global total riverine DOC flux was revised to 0.19 Pg C yr⁻¹.

Regarding DOC removal in the rest of the world's estuaries, Amon and Benner [41] estimate ~10% removal of the riverine DOC in Rio Negro, the largest left tributary of the Amazon River. Raymond and Bauer [42] report that ~10% of riverine DOC in the York River estuary is removed by bacteria. Moran *et al.* [43] report a removal rate of river DOC ranging from 1.7 to 17.7% in five estuaries of the Southeastern United States. To obtain a conservative estimate, if we further adopted an average removal rate of 10% for estuarine DOC beyond the Arctic based on the above literature, we obtained an estimate of global river DOC flux of 0.17 Pg C yr⁻¹, which is at the lower end of prior estimates.

DOC influxes from submarine groundwater discharge

In addition to surface water flow into the ocean, there has been increasing evidence pointing towards submarine groundwater discharge (SGD) into the coastal ocean. However, this groundwater contribution is difficult to quantify, partly because there are very limited DOC data from the subterranean estuaries where groundwater mixing with seawater occurs. Most available DOC concentrations in coastal groundwater have been determined in North America [44–50], and only a few studies focus on Africa [51,52], Europe [53,54] and Australia [55]. Since the average DOC is close (within the bounds of error estimation) from the different continents above, we estimated that the global average DOC was $5.9 \pm 2.5 \text{ mg L}^{-1}$ in groundwater. Given the global terrestrially derived fresh SGD ranges from 5 to 10% of the global river rate [56,57], SGD delivers 11- $22 \text{ Tg C yr}^{-1} \text{ DOC}$ to the coastal ocean globally.

Changes in riverine DOC fluxes into the ocean According to Dai et al. [15"], global river discharge between 1948 and 2004 has declined at a rate of 6.96 $\text{km}^3 \text{ yr}^{-1}$ during the past 57 years. This decrease in discharge resulted in an accumulative DOC flux decrease of only $\sim 1\%$. However, the change in discharge has not been uniform between ocean basins. River discharge into the Arctic has been changing at an increasing rate of $8.20 \text{ km}^3 \text{ yr}^{-1}$ while the rate has been declining at a rate of 1.64, 2.49 and 9.40 km³ yr⁻¹ into the Atlantic, Indian and Pacific Oceans [15^{••}]. These changes in discharge resulted in an accumulative DOC flux increase of 12.8% to the Arctic, but a decrease by 0.5, 3.1 and 5.9% to the Atlantic, Indian and Pacific Oceans. Because of the difference in river end-member DOC concentration and in DOC compositions in the rivers from different latitudes, changes in heterogenic loading would impact the carbon budgeting and cycling in the coastal ocean and ocean basin on a global scale. Moreover, such redistribution could have a significant influence in the Arctic, which is particularly prone to be increasingly vulnerable to climate change.

In addition to the observed fresh water redistribution among ocean basins, there is also evidence for changes in river DOC concentrations. For example, DOC concentrations in 22 UK upland waters have increased by an average of 91% during the last 15 years [58]. Increases have also occurred elsewhere in the UK, northern Europe and North America. A range of potential drivers of these trends are believed to be temperature, rainfall, acid deposition, land-use, and nitrogen and CO₂ enrichment [58]. Clair et al. [59] suggest that DOC export from basins in Canada might increase by 14% with a doubling in atmospheric CO₂. An additional factor rarely addressed in rivers is direct loading from urban and industrial sources [30,60]. Concentrations of DOC in New York's Hudson River have doubled over the past 16 years, implying a substantial increase in net movement of organic carbon from the watershed to New York Harbor and Bight [61]. Unfortunately, such changes at the global scale remain difficult to assess.

Concluding remarks

By using the most up-to-date river discharge values and the largest river DOC data set, this study provided an updated riverine DOC flux into the coastal ocean of 0.17 Pg C yr⁻¹. This falls at the lower end of prior estimates. At the same time, we saw a trend of river DOC increase in at least some terrestrial ecosystems, likely caused by rising temperature and atmospheric CO₂, which warrants further attention, particularly in the context of the oceanic organic carbon pool and microbial activities therein.

Spatially distributed fluxes have been presented reflecting the partitioning of organic loading into different oceanic and coastal regimes. With the identified hydrological circulation, such repartitioning might have significant implications for oceanic biogeochemistry.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.cosust.2012.03.003.

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