Reviews and syntheses: Anthropogenic perturbations to carbon fluxes in Asian river systems – Concepts, emerging trends, and research challenges

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Abstract. Human activities are drastically altering water and material flows in river systems across Asia. These anthropogenic perturbations have rarely been linked to the carbon (C) fluxes of Asian rivers that may account for up to 40– 50% of the global fluxes. This review aims to provide a conceptual framework for assessing human impacts on Asian river C fluxes, along with an update on anthropogenic alterations of riverine C fluxes. Drawing on case studies conducted in three selected rivers (the Ganges, Mekong, and Yellow River) and other major Asian rivers, the review focuses on the impacts of river impoundment and pollution on CO₂ outgassing from the rivers draining South, Southeast, and East Asian regions that

- 25 account for the largest fraction of river discharge and C exports from Asia and Oceania. Critical examination of major conceptual models of riverine processes against observed trends suggests that to better understand altered metabolisms and C fluxes in "anthropogenic land-water-scapes", or riverine landscapes modified by human activities, the traditional view of the river continuum should be complemented with concepts addressing spatial and temporal discontinuities created by human activities such as river impoundment and pollution. Recent booms in dam construction on many large Asian rivers pose a
- 30 host of environmental problems, including increased retention of sediment and associated C. A small number of studies that measured greenhouse gas (GHG) emissions in dammed Asian rivers have reported contrasting impoundment effects: decreased GHG emissions from eutrophic reservoirs with enhanced primary production vs. increased emissions from the flooded vegetation and soils in the early years following dam construction or from the impounded reaches and downstream estuaries during the monsoon period. These contrasting results suggest that rates of metabolic processes in the impounded

- 35 and downstream reaches can vary greatly longitudinally over time, as a combined result of diel shifts in the balance between autotrophy and heterotrophy, seasonal fluctuations between dry and monsoon periods, and a long-term change from a leaky post-construction phase to a gradual C sink. The rapid pace of urbanization across southern and eastern Asian regions has dramatically increased municipal water withdrawal, generating annually 120 km³ of wastewater in 24 countries, which comprises 39% of the global municipal wastewater production. Although the municipal wastewater constitutes only 1% of
- 40 the renewable surface water, it can disproportionately affect the receiving river water, particularly downstream of rapidly expanding metropolitan areas, resulting in eutrophication, increases in the amount and lability of organic C, and pulse emissions of CO₂ and GHGs. In rivers draining highly populated urban centers, lower reaches and tributaries tended to exhibit higher levels of organic C and the partial pressure of CO₂ (pCO₂) than less impacted upstream reaches and eutrophic impounded reaches, often plagued by frequent algal blooms and pulsatile CO₂ emissions from urban tributaries delivering
- 45 high loads of wastewater. More field measurements of pCO₂, together with accurate flux calculations based on river-specific model parameters, are required to provide more accurate estimates of GHG emissions from the Asian rivers that are now underrepresented in the global C budgets. The new conceptual framework incorporating discontinuities created by impoundment and pollution into the river continuum needs to be tested with more field measurements of riverine metabolisms and CO₂ dynamics across variously affected reaches to better constrain altered fluxes of organic C and CO₂
- 50 resulting from changes in the balance between autotrophy and heterotrophy in increasingly human-modified river systems across Asia and other continents.

Key words: Asian rivers, carbon dioxide outgassing, dissolved organic carbon, greenhouse gases, particulate organic carbon, river impoundment, riverine carbon flux, urbanization, water pollution

55 1 Introduction

Inland waters play a pivotal role in the global carbon (C) cycle by storing, transporting, or transforming inorganic and organic C components along the hydrologic continuum linking the land and oceans (Kempe, 1982, 1984; Cole et al., 2007; Battin et al., 2009). Recent syntheses have provided greater estimates for the riverine transport of dissolved organic C (DOC) and particulate organic C (POC) and the exchange of CO₂ between the atmosphere and inland waters than the previous studies (Raymond et al., 2013; Regnier et al., 2013; Wehrli, 2013; Ward et al., 2017). Monitoring data are sparse for many river systems in Asia and Africa, leaving many blind spots in global syntheses of riverine C transport and emission. Although Asian rivers have been estimated to account for up to 40–50% of the global inorganic and organic C fluxes from the land to the oceans (Degens et al., 1991; Ludwig et al., 1996; Schlünz and Schneider, 2000; Dai et al., 2012), the lack of high quality data and poor spatial coverage have constrained our ability to estimate the contributions of Asian river systems

to the global riverine C fluxes in general and CO₂ outgassing in particular (Schlünz and Schneider, 2000; Lauerwald et al.,

2015; Li and Bush, 2015). For instance, obtaining pCO_2 data measured in Southeast Asian rivers was suggested as a top priority to reduce the large uncertainty in estimating the global riverine CO₂ outgassing (Lauerwald et al., 2015). The release of C from anthropogenic sources in rapidly urbanizing watersheds around the world increases the uncertainty of the current global riverine C flux estimates (Regnier et al., 2013). Concurrent anthropogenic perturbations to the river

- 70 systems, including eutrophication, altered sediment regimes, and increased water residence time in impounded rivers, can significantly change the riverine processing of organic matter (OM) and greenhouse gas (GHG) outgassing (Stanley et al., 2012; Regnier et al., 2013; Crawford et al., 2016). Many streams and rivers across Asia are highly polluted by agricultural runoff and domestic and industrial wastewater, with their water quality often exhibiting large seasonal variations associated with regional monsoon rainfall regimes (Park et al., 2010; Park et al., 2011; Evans et al., 2012, Bhatt et al., 2014). Although
- 75 enhanced lability and mineralization of organic C have been observed in streams and rivers draining urbanized watersheds (Hosen et al., 2014; Kaushal et al., 2014), little is known about organic C export and CO₂ outgassing from streams and rivers draining rapidly urbanizing watersheds in developing Asian countries (Bhatt et al. 2014). A few recent studies conducted in the metropolitan areas of China and Korea have suggested that GHG emissions from polluted waterways carrying urban runoff and wastewater treatment plant (WWTF) effluents may be underappreciated as sources of GHGs (Wang et al., 2017;
- 80 Yoon et al., 2017). Despite recent booms in the construction of large dams on many large rivers across the region, little attention has been paid to impoundment effects on GHG emissions (Chen et al., 2009; Hu and Cheng, 2013). Considering the role of dams in storing huge amounts of sediment and organic C (Syvitski et al., 2005; Maavara et al., 2017), a mechanistic understanding of GHG emissions from impounded rivers can provide more insights into the anthropogenic perturbations to the C fluxes of the dammed river systems.
- This review aims to provide a conceptual framework for assessing human impacts on riverine C fluxes and an update on major anthropogenic perturbations affecting C fluxes in Asian river systems, focusing on the impacts of water pollution and river impoundments on riverine CO₂ dynamics in South (S), Southeast (SE), and East (E) Asian regions that account for the largest fraction of river discharge and C exports from Asia and Oceania (Fig. 1). An important goal was to integrate various concepts of riverine biogeochemical processes into a conceptual framework for assessing human impacts on the riverine C
- 90 fluxes in human-modified river systems. Given the pace and wide-ranging impacts of urbanization and river impoundments, the traditional view of the river continuum developed for natural streams and rivers (Vannote et al., 1980) needs to be revised to reflect altered regimes of riverine metabolic processes and material fluxes. We compared reported values of pCO_2 that had been either measured or estimated for major river basins in three Asian regions, including the Ganges, the Mekong, and the Yellow River as representative systems of three regions, to assess the current status. We also compared pCO_2 values
- among different components of the river basin (mainstem, headwater, tributary, and impoundment) to examine how water pollution and impoundments alter riverine metabolic processes and CO_2 emissions. Many of the reported values have been estimated from pH and alkalinity data available from the literature and water quality databases such as GLORICH (Global River Chemistry Database; Hartmann et al., 2014). Considering potential overestimations of water *p*CO₂ associated with organic acid contributions and increased sensitivity to alkalinity in acidic, organic-rich waters with low carbonate buffering

100 (Abril et al., 2015), we provided methodological details for the calculated pCO_2 values, if the cited references considered these pH and alkalinity effects. This review and ensuing synthesis efforts are expected to provide scientifically robust conceptual frameworks and data that are required for a better understanding of how human-induced perturbations in rapidly urbanizing watersheds across Asia transform riverine metabolic processes and C fluxes away from the 'natural' states assumed in the traditional river continuum model.

105 2 The geographical scope, global implications, and emerging regional trends of Asian river systems

Global syntheses of riverine C fluxes have been based on monitoring data available for a limited number of large rivers (e.g., Degens et al., 1991; Ludwig et al., 1996). We referred to these previous syntheses and a more recent synthesis of global river discharge (Milliman and Farnsworth, 2011) to scope the geographical extent of Asian river systems. We followed the continental categories used by Milliman and Farnsworth (2011), namely Asia and Oceania demarcated on Fig. 1, but did not

- 110 consider Arctic rivers in Russia and rivers in Australia and New Zealand. This review focuses on S, SE, and E Asian regions where river systems are commonly affected by increasing human impacts and for which data are available to address major review themes. Ten large rivers of three Asian regions (Fig. 1) belong to top 30 global rivers based on discharge (Raymond and Spencer, 2015), 32 rivers included in a previous global synthesis of riverine C fluxes (Ludwig et al., 1996), or 34 rivers with basin areas greater than 500,000 km² (Milliman and Farnsworth, 2011). The river systems in these Asian regions share
- 115 some common hydrologic and demographic features (Table 1), including a large seasonality in discharge, high population densities (80–513 km⁻²) compared to the global mean (70 km⁻²), and the wide range of per-capita annual discharge (23–8,594 m³ yr⁻¹ person⁻¹ vs. the global mean: 4,901 m³ yr⁻¹ person⁻¹). Large seasonal variations in precipitation and runoff associated with Asian monsoon systems play a critical role in hydrologically mediated riverine processes including those affecting C fluxes (Park et al., 2010). Recent demographic changes including an unprecedented rapid growth in population may cause
- 120 increasingly severe perturbations to water and material flows along the rivers that are not only regulated by dams and but also polluted by urban sewage and agricultural runoff.

According to an earlier estimation (Degens et al., 1991), Asian rivers account for up to 35%, 50%, and 39% of the global discharge, total organic C (TOC) export, and dissolved inorganic C (DIC) export. Schlünz and Schneider (2000) provided a lower estimate for TOC export by Asian rivers (175. 2 Tg C yr⁻¹; 40% of the global TOC flux). Ludwig et al. (1996)
provided separate estimates for the export of DOC (69.01 Tg C yr⁻¹) and POC (76.40 Tg C yr⁻¹) by Asian rivers, which represented 34% and 44% of the corresponding global fluxes, respectively. More recent syntheses of the published data have corroborated the quantitative importance of Asian rivers in the global fluvial C fluxes (Dai et al., 2012; Huang et al., 2012;

Galy et al., 2015). As indicated by the lower ratio of DOC to POC (0.9) compared to other regions with the ratio exceeding 1, many Asian rivers draining the erosion-prone mountainous terrain deliver more POC than DOC, particularly during the

130 monsoon period (Ittekkot et al., 1988; Ludwig et al., 1996). Monsoonal increases in discharge and POC can have either a positive effect on riverine pCO_2 levels through enhanced soil flushing of DIC and/or in-stream organic C biodegradation or a negative effect caused by dilution, as observed in such turbid Asian rivers as the Pearl (Yao et al., 2007), Yangtze (Li et al.,

2012), and Mekong (Li et al., 2013). Constraining differential monsoon effects on the fluxes of DOC, POC, and DIC including CO_2 represents a key challenge in evaluating the contribution of Asian rivers to the global riverine C fluxes.

- 135 Previous syntheses based on a small number of data sets collected in several large Asian rivers during the 1970s and 1980s provide only limited information when we assess effects of "on-going" environmental changes on riverine C fluxes. Therefore, this review aims to provide an update of Asian river C fluxes, focusing on river impoundment and pollution as two of the most important environmental changes affecting river systems across three target Asian regions. There have been few systematic assessments of effects of impoundments and water pollution on the C fluxes of Asian rivers (Sarma et al.,
- 140 2011; Ran et al., 2014; Li and Bush, 2016). For example, a cascade of dams constructed along the upper Mekong River since the 1990s have been implicated to cause a wide range of downstream impacts including decreases in water and sediment flow (Li and Bush, 2016). Although it is expected that declining sediment flux can significantly alter POC and associated C fractions along downstream reaches, little is known about impoundment effects on the fluxes of POC, DOC, DIC, and CO₂ in the upper and lower Mekong River. This lag between the real-time environmental changes and scientific assessments
- 145 based on outdated data is quite surprising, given the magnitude and pace of the environmental changes occurring across Asia. Deforestation and associated peatland drainage in tropical areas represent another important, but rarely explored topic with regard to CO₂ outgassing from Asian rivers (Baum et al., 2007; Wit et al., 2015). A recent study suggested that peatland drainage could enhance organic matter degradation in the coastal peatlands and organic rich soils of Southeast Asian lowland areas and islands, increasing CO₂ outgassing from the rivers draining the affected areas (Wit et al., 2015). However, this
- 150 issue cannot be addressed in detail here, because only a few studies have been conducted in Indonesia and Malaysia.

3 Conceptual framework for understanding interactive effects of changing land-water-scape and climate on riverine C fluxes

Human-induced land changes, as manifested in agricultural lands and urban areas, drive changes in biogeochemical cycles and climates, with altered terrestrial biogeochemical cycles often leading to pollution in downstream aquatic systems (Grimm et al., 2008). As a consequence of global urbanization, human influences are pervasive across the interacting terrestrial and aquatic patches of riverine landscapes or riverscapes (Allan, 2004; McCluney et al., 2014). To emphasize dominant human influences on connectivity and interactions among terrestrial and aquatic patches of the riverine networks, we term these anthropogenically modified riverscapes "anthropogenic land-water-scapes". Compared to the previous use of the term "land-waterscape" focusing on terrestrial-aquatic boundary conditions in urbanized watersheds (Cadenasso et al., 2008), our use is more general and inclusive, covering longitudinal linkages between less or more modified up- or

- downstream reaches. Rivers dominated by effluents of wastewater treatment plants (WWTPs) provide an example of how human activities, through water withdrawal and wastewater generation, modify flows of water and materials across this anthropogenic land-water-scape. In the case of rivers draining arid areas, WWTP effluents can not only increase river flow
- 165 but also provide a source of water feeding into habitats for various aquatic organisms along downstream reaches (Luthy et al.,

2015). Rapid, concurrent changes in land use and river flow and chemistry make Asian rivers a perfect test bed for exploring how human-induced perturbations alter hydro-biogeochemical cycles across the components of these anthropogenic land-water-scapes.

New conceptual templates can build on some existing concepts that have been used to explain natural riverine processes.

- 170 Above all, the river continuum from headwaters to mouth has been one of the most widely used concepts to represent longitudinal connectivity in river ecosystem structure and function over the last five decades (Vannote et al., 1980; Webster, 2007). The original river continuum concept envisaged gradual and continual changes in OM composition and metabolic rates in correspondence to downstream variations in environmental conditions and biotic communities along the river (Vannote et al., 1980; Fig. 2). Recent biogeochemical studies based on the river continuum concept include those of OM
- 175 chemical diversity (Mosher et al., 2015) and biodegradability (Catalán et al., 2016), and riverine CO₂ outgassing (Hotchkiss et al., 2015). A prevailing idea underlying these approaches has been the selective degradation of labile components of OM during transit across the continuum, which has been successful in explaining the critical role of water retention time for the downstream evolution of the composition and biodegradability of DOM in the river systems with a relatively high proportion of natural lakes and/or low levels of anthropogenic perturbations (Koehler et al., 2012; Weyhenmeyer et al., 2012; Mosher et al., 2012;
- 180 al., 2015; Catalán et al., 2016). According to the reactivity continuum model, the composition of DOM becomes dominated gradually by highly degraded compounds as a result of prolonged exposure of DOM to biodegradation and photodegradation, resulting in a downstream decline in DOM reactivity (Koehler et al., 2012; Catalán et al., 2016).
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Despite the wide use of the river continuum in studying various riverine processes, it has been criticized for overlooking an increasingly recognized reality that specific rivers are often divided into discrete segments that are hierarchically nested in a

- 185 river network (Townsend, 1996; Poole, 2002). Discrete segments along a river network can occur as a result of "abrupt transitions between adjacent segments with dissimilar physical structure" within the hierarchically nested river network (Poole, 2002). These abrupt transitions between discrete segments can occur temporarily, as illustrated by seasonal variations in water connectivity (Casas-Ruiz et al., 2016). As depicted in Fig. 2, examples of human-induced discontinuities include those created by dams built on regulated rivers (Ward and Standford, 1983) and pollution-induced perturbations of the
- 190 production-respiration balance in the eutrophic river (Kempe, 1984; Garnier and Billen, 2007). Because the river continuum concept was originally proposed as a template for integrating physical environments and biological processes of "natural, unperturbed stream ecosystems" (Vannote et al., 1980), it has limitations in explaining discontinuities in fluvial processes and biogeochemical fluxes, which might be accentuated in many anthropogenically modified Asian river systems.
- River impoundments and water withdrawal alter not only the rates of runoff and sediment transport (Syvitski et al., 2005) but also aquatic primary production and its effects on OM biodegradation (Stanley et al., 2012). In response to disturbance events, unregulated rivers tend to reset physical and ecological conditions toward the pre-disturbance state. However, river impoundment induces long-lasting perturbations to those conditions along the distance up- or downstream of a dam, termed 'discontinuity distance' (Ward and Standford, 1983; Stanford and Ward, 2001). This discontinuity distance was originally proposed as part of the 'serial discontinuity concept', which states that stream regulation by multiple dams results in "an

- 200 alternating series of lentic and lotic reaches" (Ward and Stanford, 1983). According to this concept, stream regulation by dams can induce disturbances to the gradual processes envisaged in the river continuum concept, shifting a given physical or biological parameter longitudinally (Ward and Stanford, 1983). For example, Vannote et al. (1980) envisaged that parameters such as the ratio of production to respiration (P/R) and diel temperature difference (Δ T) would exhibit a specific longitudinal pattern shown in Fig. 2. Stream regulation can shift this longitudinal pattern along the discontinuity distance.
- 205 Although serial discontinuity concept has been a useful framework for assessing anthropogenic impacts on regulated lotic systems, its presuppositions including no disturbances other than impoundment (Ward and Stanford, 1983) limit its application to investigating other environmental stresses than impoundments, such as high levels of organic pollutants and nutrients observed in many Asian rivers receiving untreated sewage and urban runoff.
- Kaushal and Belt (2012) proposed an urban watershed continuum framework that recognizes a continuum linking engineered and natural hydrologic flowpaths across the urbanized watershed. From the perspective of spatial disconnection within the hierarchically nested river network (Poole, 2002), this urban watershed continuum is actually "a continuum with discontinuities" (*sensu* Poole, 2002), in which the natural land-water hydrologic connectivity common in low-order streams is replaced by urban structures such as sewers and stormwater drains. The lateral transfer of water and associated materials via networks of engineered urban structures not only creates departures from the natural patterns or "discontinuities" in the
- 215 hydrologic paths across the terrestrial-aquatic interface, but also exert extraordinary impacts on downstream transport and transformations of OM and nutrients (Paul and Meyer, 2001; Allan, 2004; Garnier and Billen, 2007; Lookingbill et al., 2009; Kaushal and Belt, 2012). As Hynes (1975) emphasized the importance of the terrestrial-aquatic connectivity in headwater systems by saying that "the valley rules the stream, human-induced changes in the watershed would have large cascading effects on the structure and function of stream ecosystems. In human-modified river systems, the "valley" is often separated
- 220 from the stream or replaced by engineered structures that release pulses of water and materials, creating abrupt transitions across the land-water interface and stream segments (Fig. 2). Urban structures across the land-water interface can also result in pulsatile flows of water and materials as a combined consequence of increased runoff from the impervious urban surface and discharges from WWTPs, stormwater drainages, and combined sewer overflows (Paul and Meyer, 2001; Garnier and Billen, 2007; Kaushal and Belt, 2012). Although wastewater can bring pulses of OM and nutrients to the receiving urban
- 225 water systems, its impact on riverine metabolism and CO₂ outgassing has rarely been investigated in Asian river systems except for a few exploratory studies (Guo et al., 2014; Yoon et al., 2017). Human-modified river networks often lack dynamic movements and flow adjustment and are therefore limited in their ability to buffer against disturbances such as floods and water stress (Palmer et al., 2008). Given the large seasonality inherent in the monsoon climate, anthropogenic land-water-scapes forming on monsoonal Asian river systems might be particularly
- 230 vulnerable to climatic variability and extremes, as exemplified by strengthened flashy storm responses of sediment and C export from erosion-prone mountainous watersheds during extremely wet monsoon periods (Park et al., 2010; Jung et al., 2012). Climate models have suggested that perturbations in the global water cycle accompanying climatic warming can increase river discharge in many parts of the world (Milly et al., 2005). Although increases in river discharge have been

detected for some large basins over the last century (Labat et al., 2004), globally no consistent pattern has been established.

- For example, cumulative discharge from many mid-latitude rivers including rivers draining arid regions of Asia have decreased substantially as a result of concurrent changes in precipitation and anthropogenic perturbations such as damming, irrigation, and inter-basin water transfers (Milliman et al., 2008). While discharge from most of Asian rivers except Siberian rivers and the Brahmaputra has declined, the most striking decrease exceeding –50% was observed for the Indus and Yellow River (Milliman et al., 2008). In many dammed Asian rivers, observed decreases in discharge and sediment transport might
- 240 be largely explained by increasing river impoundments and water diversion (Milliman et al., 2008; Li and Bush et al., 2016). However, recent increases in the frequency and intensity of extreme precipitation events observed in many parts of Asia (Min et al., 2011) suggest that potential changes in monsoon rainfall regimes as a consequence of climate change can amplify seasonal and year-to-year variations in discharge and the transport of sediment and C even in dammed river systems. Therefore, predicting future changes of riverine C fluxes in increasingly human-modified Asian river systems would require
- a better understanding of the complex interplay between anthropogenic perturbations and the concurrent climate change.

4 Contrasting effects of river impoundment on organic C transport and CO₂ emission

According to a recent estimate based on the Global Reservoir and Dam database (GRanD), there may exist about 16.7 million reservoirs larger than 0.01 ha globally, with a combined storage capacity of 8069 km³ (Lehner et al., 2011). Out of 6862 dams registered in GRanD with a total storage capacity of 6197 km³, 1906 dams located in Asia (excluding Middle East Asia and western Russia) store 1625 km³ water, accounting for 26% of the global storage capacity. AQUASTAT, a global water information system operated by Food and Agriculture Organization (FAO), provides a similar estimate for the reservoir storage capacity of southern and eastern Asia: 1325 km³ (Table 2; FAO, AQUASTAT). Over the last decades, rivers across Asia have been increasingly impounded by dams of various type and size and a recent boom in constructing hydroelectric dams is posing an unprecedented challenge for the sustainable management of the affected river basins (Grumbine et al., 2012; Winemiller et al., 2016). River impoundments not only affect downstream flows but also disrupt the ecological and biogeochemical connectivity of rivers (Lehner et al., 2011; Winemiller et al., 2016; Maavara et al., 2017). A

- growing number of dams have been decreasing both river flow and sediment fluxes, sequestering over 100 billion metric tons of sediment and 1 to 3 billion metric tons of C in reservoirs constructed over the last 50 years (Syvitski et al., 2005).
- 260 Many Asian rivers, such as the Indus, Yangtze, and Yellow, have seen largest reductions in sediment export to the oceans compared to the pre-dam era (Syvitski et al., 2005). Therefore, investigating altered rates of C storage and losses in dammed Asian rivers is crucial for a better understanding of human impacts on global riverine C fluxes.

The Yellow River in northern China provides an excellent example of basin-scale impoundment impacts on the sediment and C transport (Fig. 3). The sediment load in the Yellow River peaked during the period 1800–1950 following a millennium of

aggravating soil erosion in the Loess Plateau, but dam construction and other human activities to control soil erosion have reduced the annual sediment flux by 90% over the last 60 years (Chen et al., 2015; Wang et al., 2015). Particularly, silt check dams and reservoirs, along with hillslope soil conservation measures such as terrace farming, made large contributions to the observed reductions. More than 110,000 silt check dams have been constructed in the Loess Plateau since the 1950s, trapping approximately 21 billion tons of sediment in the reservoirs (Zhang et al., 2016). A conservative estimate indicates

- 270 that the rate of annual POC trapping in more than 3,000 large dams (excluding 110,000 silt check dams) within the Yellow River basin can amount to 3.3-4.3 Tg C yr⁻¹ (1 Tg = 10^{12} g; Zhang et al., 2013; Ran et al., 2014), which is similar in magnitude to the total organic C export to the Bohai Sea (4.1 Tg C yr⁻¹; Ran et al., 2014; Fig. 3). Furthermore, as an important attempt to control soil erosion, the Chinese government initiated the largest ever revegetation program in history called the "Grain for Green Project" from the late 1990s. Implementation of this project has made an additional contribution
- 275 to the decreasing trend of sediment flux (Wang et al., 2015), with far-reaching impacts on soil organic C stocks and the riverine C fluxes (Feng et al., 2013). A great number of new silt check dams up to 163,000 are being planned to be built on the Loess Plateau through 2020 (Zhang et al., 2016), so dams and other erosion control projects will continue to have significant impacts on sediment and associated C dynamics in the Yellow River basin in the future.
- It remains an important research question whether POC trapped in ever-growing reservoir sediments within the Yellow River basin would function as a sink or source of CO₂ for the atmosphere (Zhang et al., 2013; Ran et al., 2015a, 2017a). As illustrated by the relatively high estimated rate (27%) of CO₂ emission from the POC eroded from the entire Yellow River basin (Ran et al., 2014; Fig. 3), the organic C trapped in reservoir and stream sediments can become an important source of CO₂ under favorable conditions that would accelerate the rate of biodegradation of POC during the fluvial transport and sediment storage. Although the rate of CO₂ evasion from the water surface can increase in the impounded river reaches as
- longer residence times tend to create favourable environments for microbial biodegradation of organic C (Ittekkot et al., 1985; Ran et al., 2015a), the countering effect of enhanced planktonic CO_2 uptake in the euphotic reservoir surface has rarely been compared against biodegradation (refer to the wide range of pCO_2 summarized in Table 3). Based on extensive CO_2 evasion measurements in the river-reservoir-river continuum on the Loess Plateau, Ran et al. (2017a) found that the Loess Plateau reservoirs acted as relatively small sources or even sinks of C, due largely to the significantly reduced surface
- 290 turbulence and enhanced photosynthesis. Compared to the standing waters of the reservoirs with enhanced primary production, rapidly flowing waters along both the upstream and downstream reaches were larger C sources for the atmosphere, exhibiting much higher pCO_2 levels and faster flow velocities providing favorable conditions for an efficient gas evasion from the aqueous boundary layer. Liu et al. (2016) also observed over 60% decreases in pCO_2 along the eutrophic impounded reaches of the Three Gorge Reservoir in the Yangtze River, but they argued that enhanced primary production in
- 295 the impounded reach would play a rather temporary and minor role in controlling pCO_2 dynamics compared to the predominant influence of allochthonous C. The contrasting impoundment effects observed in the Yellow River and other Chinese rivers suggest that a basin-wide assessment of impoundment impacts on sediment C storage and CO₂ emissions should take into consideration concurrent changes in primary production and organic matter biodegradation.

In the Lancang River (the upper Mekong River located in China), Li et al. (2013) observed dramatic increases in the abundance of phytoplankton and a shift of the algal community toward *Chlorophyta* and *Cyanophyceae*, following the

construction of cascade dams since 1995. In a recent study that compared the emission rates of CO_2 and CH_4 across the upper riverine reach and six cascade dams along the Lancang River, Shi et al. (2017) found that gas emission rates, particularly those of CH_4 , were highest in the most upstream and second newest dam and that % organic C in the reservoir bottom sediment had decreased with the increasing age of the dams. These results suggest that favourable conditions created

305 by river impoundments, such as increased water temperature and retention time, can stimulate OM processing in both the impounded water and trapped sediments, at least in the short term following the construction of dams on the high-POC mountainous rivers such as the Lancang.

Most major Indian rivers have been impounded by dams of various type and size to meet domestic and industrial water demands during the dry period. Because monsoonal rivers draining the Indian subcontinent account for the largest share of

- 310 the POC export by the Asian rivers (Ludwig et al., 1996; Galy et al., 2015), altered discharges and sediment fluxes of these dammed rivers can have significant implications for the global riverine export of sediment and POC (Ittekkot et al., 1985; Krishna et al., 2015). However, effects of large dams on riverine C fluxes including CO₂ have been studied only in a few dammed rivers such as the Godavari (Sarma et al., 2011; Prasad et al., 2013; Table 3). According an earlier study conducted in the Krishna, dam construction had decreased the sediment load from 67.7 Tg yr⁻¹ measured at the upper reach to 4.11 Tg
- 315 yr⁻¹ at the river mouth (Ramesh and Subramanian, 1988). Although many large dams and barrages have been constructed on the mainstem and tributaries of the Ganges, there has been no systematic investigation of GHG emissions from the impounded reaches, except for a few unpublished measurements (Table 3). Because of this paucity of monitoring data, GHG emissions from impounded rivers add a considerable uncertainty to the estimates of GHG emissions from the inland water systems in the Indian subcontinent. For instance, Panneer Selvam et al. (2014) extrapolated their flux measurements at 45
- 320 water bodies in South India to estimate CO_2 and CH_4 emissions from the entire India's inland waters at 22.0 Tg CO_2 yr⁻¹ and 2.1 Tg CH_4 yr⁻¹, respectively. While they provided estimates of 2.37 Tg CO_2 yr⁻¹ and 0.33 Tg CH_4 yr⁻¹ for the reservoirs and barrages, a follow-up study offered larger estimates amounting to 3.08 Tg CO_2 yr⁻¹ and 6.27 Tg CH_4 yr⁻¹ by considering additional literature data on large rivers in northern India and reservoir downstream fluxes through spillways and turbines (Li and Bush, 2015). This example illustrates the importance of adequate spatial coverage and downstream impacts for refining
- 325 regional-level estimates of GHG emissions from various impoundments. A significant reduction in the primary production observed in the Godavari River was ascribed to the removal of nitrogen and phosphorus by several dams constructed within the basin (Das, 2000). Ramesh et al (2015) reported increasing retention of particulate OM in the dams and reservoirs of the Godavari and Krishna. However, other studies conducted in the Godavari estuary have found that during the peak discharge periods in the monsoon season, the estuary receiving discharge waters
- 330 from an upstream dam exhibited extraordinarily high levels of pCO_2 up to 33,000 µatm compared to the dry season values lower than 500 µatm, presumably due to enhanced bacterial decomposition of the organic C released from the upstream dam in the highly eutrophic estuary (Sarma et al., 2011, Prasad et al., 2013). On the other hand, less rainfall during dry years can also result in large downstream impacts of impoundments through an increased production of labile OM by freshwater algae in the upstream dam and algal blooms in the estuary (Pradhan et al., 2014) It demands further research to establish how

335 seasonal and inter-annual variations in climatic and trophic conditions in dammed Indian rivers alter the balance between autotrophy and heterotrophy and hence CO₂ emissions along the "discontinuous" river-reservoir-estuary continuum as found in the Godavari basin.

In accordance with the serial discontinuity concept (Ward and Standford, 1983; Fig. 2), multiple dams constructed on large Asian rivers such as the Mekong and Yellow River create standing water conditions that may shift stream metabolisms and

- 340 pCO_2 dynamics from the patterns observed for freely flowing reaches. The observed contrasting impoundment effects on CO₂ emission across different Asian river systems might have resulted from an interplay between planktonic CO₂ uptake, organic matter biodegradation, and sediment C sequestration (Liu et al., 2016; Maavara et al., 2017). The balance between the competing processes affecting the actual level of pCO_2 in reservoir waters may change not only seasonally (Prasad et al., 2013; Liu et al., 2016) but also with the increasing age of dams (Barros et al., 2011). While large pulses of GHGs may be
- released from the flooded vegetation and soil OM during the initial flooding phase (Abril et al., 2005; Chen et al., 2009; Hu and Cheng, 2013; Deshmukh et al., 2016, 2017), sedimentation can accumulate a growing amount of C in reservoir sediments, greatly decreasing the rate of CO_2 release from aging reservoirs (Barros et al., 2011). Large pulse emissions of CO_2 and CH_4 have been measured in the years following the construction of the Three Gorges Dam on the Yangtze River (Chen et al., 2009) and the Nam Theun 2 on the large tributary feeding into the middle reach of the Mekong River
- 350 (Deshmukh et al., 2016, 2017). A recent report on drought-enhanced emissions of GHGs in an old hydroelectric reservoir in Korea suggested that stochastic emissions during extreme climatic events can reverse the trend of declining C emissions from aging reservoirs by the offsetting effect of extreme events on the C accumulated in reservoir sediments over time scales of years to decades (Jin et al., 2016). The paucity of pCO_2 measurements in dammed Asian rivers (Table 3; Fig. 4) does not allow for any generalization of long-term impoundment effects on sediment C storage and CO₂ emissions, demanding more
- 355 long-term investigations of seasonal and year-to-year variations in metabolic processes and pCO_2 levels across a wide range of impounded inland water systems.

5 Effects of water pollution on riverine metabolisms and CO₂ emissions

Across Asia rapidly urbanizing river basins are highly polluted with poorly treated or untreated wastewater. Using AQUASTAT data, Evans et al. (2012) estimated the annual wastewater generation in Asia around the year 2000 at 142 km³, of which only an estimated 33–35% was treated before being discharged to streams and rivers. We used the latest data available on the AQUASTAT webpage to provide more up-to-date estimates of water withdrawal and wastewater production, focusing on southern and eastern Asia (FAO, AQUASTAT). In 2010, the annual municipal water withdrawal in 24 southern and eastern Asian countries was 201.6 km³, accounting for 43.4% of the global municipal withdrawal (464.1 km³; Table 2).

The total volume of the municipal wastewater generated each year within urban areas of these countries was 120.2 km³. The generated wastewater included domestic, commercial, and industrial effluents, and storm water runoff, accounting for 38.6% of the global municipal wastewater production (311.6 km³). Based on the AQUASTAT and other published data, Mateo-

Sagasta et al. (2015) estimated that each year more than 330 km³ of the municipal wastewater are produced globally. Although the volume of the municipal wastewater generated in these Asian regions constitutes only \sim 0.9% of the renewable

Compared to extensive studies conducted in polluted rivers and estuaries in Europe and North America (Frankignoulle et al.,

- 370 surface water available in these regions (14,027.1 km³), both poorly treated and untreated wastewater can have disproportionately large impacts not only on the water quality and ecological integrity of downstream aquatic ecosystems (Meybeck and Helmer, 1989; Evans et al., 2012) but also on the riverine GHG emissions (Yoon et al., 2017).
- 1998; Borges et al., 2006; Hartmann et al., 2007; Borges and Abril, 2011; Griffith and Raymond, 2011; Amann et al., 2012; 375 Joesoef et al., 2015), few efforts have been made to measure pCO_2 in polluted Asian rivers, except for some large rivers and estuaries in East Asia (Zhai et al., 2005; Chou et al., 2013; Ran et al., 2015b; Yoon et al., 2017). These studies, together with a small number of studies that used water chemistry data to estimate pCO_2 levels in major Asian rivers such as the Mekong (Li et al., 2013), the Yangtze (Ran et al., 2017b), the Ganges-Brahmaputra (Manaka et al., 2015), and Indian estuaries (Gupta et al., 2009; Sarma et al., 2012), underscored the importance of anthropogenic OM and nutrients for riverine CO_2 dynamics,
- 380 particularly along lower river reaches and estuaries draining highly populated areas. When published data of pCO_2 were compared between headwater streams and tributaries feeding into the middle and lower reaches of major Asian rivers, tributary pCO_2 levels (mean: 2,118 µatm) tended be higher than those for headwaters of global rivers (mean: 1,116 µatm) and Asian rivers (mean: 973 µatm) (Table 3; Fig. 4). The ranges of pCO_2 in the three river systems reviewed in detail also differed between headwaters and lower mainstem reaches and their tributaries, displaying some river-specific patterns as
- 385 described in the following paragraphs.

With the basin-wide average pCO_2 around 2,164 (147–9,659) µatm (Table 3), the Yellow River has been evaluated as a source of CO₂ for the atmosphere (Ran et al., 2015a, b; Ran et al., 2017a; Fig. 3). The total emission of CO₂ from the basin-wide fluvial network was estimated at 4.7–7.9 Tg yr⁻¹, with >70% emitting from the tributaries draining the Loess Plateau (Ran et al., 2015b; Fig. 3). In recent decades water pollution has become an increasingly important watershed

- 390 management issue for the basin inhabited by a population > 100 million, particularly in the middle and lower reaches flanked by large industrial complexes and irrigated farmlands (Li et al., 2006; Zhang et al., 2013; Lu et al., 2015). Drainage waters from croplands containing high loads of OM have increased DOC concentrations in the middle reach of the Yellow River, while wastewater discharged from large regional population centers has been evaluated as the major source of DOC and POC in the lower reach, particularly in winter (Zhang et al., 2013). As a combined result of higher loads of pollutants
- 395 discharged from local sources to tributaries and higher flow diluting pollutant concentrations in the mainstem, tributaries appear to be more polluted than the mainstem Yellow River, exhibiting the highest levels of pCO_2 among the three compared river systems (Fig. 4). Higher concentrations of DOC and POC in more polluted lower reaches and their tributaries might lead to enhanced in-stream biodegradation of allochthonous C by labile OM fractions of anthropogenic origin, but altered rates of biodegradation and primary production have not yet been measured in any reach of the Yellow River. Along with the
- 400 question about impoundment effects on sediment C, the role of organic pollutions in riverine metabolisms and CO₂

emissions along lower reaches is crucial for understanding the fate of organic C derived from various sources in the Yellow River basin including C stocks stored in reservoir and floodplain sediments.

It is very difficult to evaluate overall pollution impacts on C dynamics of the mainstem Mekong River, as measurements of the nutrient and C cycles have been made for short reaches of the lower Mekong in Laos and Cambodia (Alin et al., 2011;

- Ellis et al., 2012; Martin et al., 2013) and in the Mekong Delta (Borges et al., 2017). The annual DIC flux of the Mekong was estimated at 3.95 Tg, with DIC and alkalinity both negatively correlated with discharge (J. Richey, unpublished data). This trend, which has also been observed in the Pearl (Zhang et al., 2007) and Yellow River (Ran et al., 2015a), has been attributed to the dominance of a weathering-based source in the dry season which is diluted by less ion-rich rainwater during the high-flow periods (Cai et al., 2008). The mean of reported *p*CO₂ values for the lower Mekong River is 1235 μatm (Table
- 410 3). Using a model based on pH and alkalinity, Li et al. (2013) reported a similar mean of pCO₂ for the Lower Mekong: 1090 (224–5,970) µatm. Alin et al. (2011) measured similar values (~1200 µatm) at eight mainstem locations in Laos and Cambodia. The seasonal trend in pCO₂ opposes the alkalinity and DIC trends, peaking in the flood season and lowest in the dry season, similar to several previous studies in tropical river systems (Sarma et al., 2011; Borges et al., 2017). The level of pCO₂ in the Lower Mekong tend to increase downstream, with an average of 812 µatm near Chiang Saen, Thailand
- 415 increasing toward 1670 μ atm in the Mekong Delta (Li et al., 2013). Potential effects of polluted tributaries on mainstem CO₂ emissions were indicated by high *p*CO₂ values approaching 2000–3000 μ atm in tributaries draining the highly populated areas such as the Tonle Sap near Phnom Penh and local tributaries feeding to the Mekong Delta (Li et al., 2013; Table 3). In Phnom Penh, a combined drainage system delivers untreated municipal wastewater and storm runoff either directly or through four natural wetlands surrounding the city used for natural purification to the Tonle Sap and Mekong (Irvine et al.
- 420 2006). Measurements of *p*CO₂ along three freshwater channels in the Mekong Delta ranged between 1,895 and 2,664 μatm during the high flow periods of December 2003 and October 2004 (Borges et al., 2017) and exceeded the range of 703–1,597 μatm observed in the upstream reach during the similar period (September–October, 2004 and 2005) by Alin et al. (2011). As suggested by Borges et al. (2017), anthropogenic pollution sources in the densely populated and cultivated areas along lower reaches may release more CO₂ and biodegradable OM compared to the upstream reach.
- 425 As summarized in Table 3, many studies of aquatic CO_2 dynamics in India have been conducted in estuaries and coastal areas (Mukhopadhyay et al., 2002; Biswas et al., 2004; Gupta et al., 2009; Sarma et al., 2012; Samanta et al., 2015), except for the secondary data of pCO_2 calculated using C system equations (Pierrot et al., 2006) and water quality data collected in various headwaters (Sarin et al., 1989; Bickle et al., 2003; Chakrapani and Veizer, 2005) and lower reaches (Manaka et al., 2015) of the Ganges-Brahmaputra. The values of pCO_2 estimated for some headwaters, lower reaches, and tributaries of the
- 430 Ganges basin (mean: 893; range: $65-2,620 \mu atm$) were relatively low compared to other Asian rivers (Table 3; Fig. 4). In a study of human impacts on C dynamics in the Cochin estuary, Southern India, Gupta et al. (2009) ascribed monsoonal *p*CO₂ increases up to 6,000 μatm to the enhanced decomposition of the OM released from anthropogenic sources upstream. A particular attention has been paid to the emission of CO₂ and CH₄ from the Indian part of the deltaic region of the Ganges-

Brahmaputra system that includes estuaries with contrasting biogeochemical features: the anthropogenically impacted

- 435 estuary of the Hooghly (the largest Indian distributary of the Ganges emptying into the Bay of Bengal) and the mangrovedominated estuaries of Sundarbans, the world's largest mangrove ecosystem (Mukhopadhyay et al., 2002; Biswas et al., 2004; Dutta et al., 2015; Samanta et al., 2015). The Hooghly estuary was found as net heterotrophic, with the fugacity of CO₂ (*f*CO₂) varying from ~400–700 µatm (Mukhopadhyay et al., 2002). On an annual scale, the Hooghly estuary acts as a source of CO₂ (–2.78 to 84.4 mmol m⁻² d⁻¹) to the atmosphere (Mukhopadhyay et al., 2002). The estuaries of Sundarbans
- 440 were reported as a source of CO₂ (314.6 mmol m⁻² d⁻¹) to the atmosphere (Dutta et al., 2015). Samanta et al. (2015) reported a large annual DIC export $[(3.1-3.7) \times 10^{12}g]$ from the Hooghly estuary to the Bay of Bengal exceeding the input of DIC through the river (freshwater). They attributed the estuarine production of DIC to some biogeochemical processes within the estuary including OM biodegradation and carbonate dissolution. Drawing on wastewater discharge and DIC concentrations, Samanta et al. (2015) estimated that direct anthropogenic sources of DIC within the Hooghly basin might account for only
- 445 2–3% of the river water DIC concentrations. It remains unanswered how much the biodegradation of organic C released from anthropogenic sources could contribute to downstream DIC generation and CO₂ emissions in the Hooghly and other Indian estuaries.

The fact that in all three river systems pCO_2 tended to be higher along lower reaches and tributaries than in headwater streams (Table 3; Fig. 4) might seem contradictory to findings of some recent global syntheses that compared pCO_2 levels

- 450 between low-order streams and rivers (Lauerwald et al., 2015; Marx et al., 2017). These syntheses assumed that the stream pCO_2 level might be determined by the relative contributions from terrestrial processes such as soil respiration and weathering and in-stream processes including biodegradation and photodegradation. It follows then that the contribution of terrestrially derived pCO_2 may overwhelm the in-stream contribution, at least in low-order streams and rivers, resulting in a general trend of downstream decreases in pCO_2 . These assumptions, together with a finding of gradual downstream decline
- 455 in the rate of the biodegradation of riverine OM due to an increasing dominance of recalcitrant components with increasing stream order and retention time (Catalán et al., 2016), are based on the river continuum concept and do not consider downstream variations in the consumption and replenishment of stream OM pools associated with enhanced algal production in impounded reaches and/or pulsatile inputs of anthropogenic OM delivered by urban streams as described in Fig. 2. In a global synthesis of data collected at 1182 sites, which did not include highly polluted river sites in Europe and the three
- 460 Asian regions reviewed here, Lauerwald et al. (2015) found a slightly higher mean pCO_2 (2,471 µatm) in streams and small rivers compared to the mean value for large rivers (2,299 µatm). As Lauwerwald et al. (2015) acknowledged, however, pCO_2 may continue to increase along lower reaches of large rivers in response to inputs of labile OM from floodplains (Abril et al., 2014) and pollution sources in croplands and urban areas (Kempe, 1982; Frankignoulle et al., 1998). Although it is very difficult to evaluate the relative contributions of autochthonous, soil-derived, and anthropogenic OM fractions to enhanced
- 465 biodegradation along lower reaches, some studies have examined the effects of domestic and industrial wastewaters on the chemical composition and lability of organic C in riverine and estuarine waters of some polluted Asian rivers (Guo et al.,

2014; Samanta et al., 2015). By comparing fluorescence excitation-emission matrices (EEMs) of DOM between the branches and tributaries of the Yangtze River estuary, Guo et al. (2014) found that labile DOM components delivered by the Huangpu River, a highly polluted tributary, exerted a disproportionately large influence on the biodegradability of DOM in the

470 Yangtze estuary. These chemical analyses, together with direct underway measurements that revealed extraordinarily high pCO₂ levels along polluted river and estuarine reaches of some Chinese rivers (Zhai et al., 2005; Chou et al., 2013; Wang et al., 2017), suggest that labile OM fractions of anthropogenic origin can boost microbial processing of the bulk riverine OM, enhancing CO₂ emissions from polluted waterways.

Unusually high pCO₂ levels observed in some eutrophic rivers and estuaries across the three Asian regions cannot be

- 475 explained by widely used metabolic continuum models that would be more relevant for rather "natural" inland water systems (Hotchkiss et al., 2015; Catalán et al., 2016). Many studies conducted in polluted European rivers reported frequent occurrences of extreme algal blooms and altered metabolic rates in the eutrophic reaches that had rarely been observed in flowing water systems under minimal to low human influences (Meybeck and Helmer, 1989; Hilton et al, 2006; Garnier and Billen, 2007). A recent report on CO₂ outgassing from a highly urbanized river system in Korea suggested a potential regime
- 480 shift in riverine metabolic processes by showing a shift in the relationship between Chl *a* and pCO_2 from the upstream reach less enriched in nutrients and CO₂ to the eutrophic downstream reach receiving highly polluted urban tributaries carrying WWTP effluents (Yoon et al., 2017; Fig. 5). This case study was conducted in a heavily impounded and populated river basin (the Han River) where the middle reach is impounded by cascade dams and the lower reach receives loads of OM and nutrients delivered by urban streams draining the Seoul metropolitan area with > 20 million population. Therefore, multiple
- 485 dams along the middle reach and pulsatile inputs of OM and nutrients along the lower reach may create discontinuities in metabolic processes and CO_2 emissions along the longitudinally connected reaches, providing an excellent example of the anthropogenic land-water-scape depicted in Fig. 2. In accordance with the findings of large spatial and seasonal variations in the balance between autotrophy and heterotrophy in eutrophic European rivers (Garnier and Billen, 2007), enhanced bacterial degradation of OM of both allochthonous and autochthonous origin in the eutrophic lower reach receiving high loads of organic pollutants might increase the level of pCO_2 substantially despite the longitudinal increase in primary
- production with widening channel toward the river mouth, shifting the regime of riverine metabolisms away from those found in the less eutrophic upstream reach (Fig. 5).

6 Summary and future research needs

495 This review identified alarming regional trends concerning dam construction booms and the rapid pace of urbanization across three reviewed Asian regions, both of which can significantly alter riverine metabolisms and C dynamics. Some recent studies have reported significant, but contrasting impoundment effects on GHG emissions and sediment C storage in dammed rivers. As illustrated by large pulse emissions of CO₂ and CH₄ in the years following the construction of the Three Gorges Dam on the Yangtze River and cascade dams and the Nam Theun 2 in the Lower Mekong River basin, flooded soils

- 500 and vegetation can become major sources of GHGs during the initial years following dam construction. Long-term changes in GHG emissions and sediment C storage might vary with dam location, initial conditions of the flooded area, and land use changes occurring within the watersheds. As summarized in Table 3, there have been only a small number of pCO_2 measurements in dammed Asian rivers and almost no study that tracked long-term changes in CO₂ dynamics in impounded reaches and downstream rivers, making it very difficult to constrain factors crucial for the spatial and temporal variations in
- 505 pCO_2 along the impounded reaches. Unlike in Europe and North America where very few large dam projects have been commissioned over the recent decades, the current booms of mega dam construction across Asia appear to induce everincreasing perturbations to riverine C fluxes, demanding more systematic assessments of impoundment impacts on riverine organic C transport and GHG emissions. Specifically, these assessments require a basin-wide examination of temporal variations in rates of the primary production and organic matter biodegradation in line with pCO_2 variations across
- 510 impounded and up- and downstream reaches along the 'discontinuity distance' (Ward and Standford, 1983) to predict how impoundments alter the balance between autotrophy and heterotrophy and hence the air-water exchange of CO₂ both in the short-term diel cycle and through seasonal and inter-annual variations. Long-term studies can also evaluate how frequent droughts associated with regional climate change can reverse the gradually decreasing GHG emissions from impounded river reaches through enhanced decomposition of the C stored in the reservoir bottom sediment.
- 515 Although rapid urbanization across Asia is aggravating eutrophication and organic pollution in many large rivers draining metropolitan areas with a limited capacity of the wastewater treatment infrastructure, the scarcity of high-quality monitoring data represents a huge challenge for a thorough assessment of the current status of riverine metabolisms and CO₂ outgassing from the impacted rivers. Some exploratory studies conducted in highly urbanized watersheds in East Asia (e.g., Wang et al., 2017; Yoon et al., 2017) have questioned whether the conventional conceptual framework perceiving riverine C fluxes as a
- 520 gradual longitudinal continuum can address large cross-scale variations and pulsatile patterns of riverine CO_2 outgassing observed in the highly modified river systems. Given the large share of the reviewed Asian regions (~40%) in the global municipal wastewater production and disproportionately poor wastewater treatment infrastructure, it remains largely unknown whether our current understanding of biogeochemical processes in urban river systems in Europe and North America can help explain idiosyncratic features of OM composition and turnover in streams and rivers contaminated with
- 525 high loads of raw sewage and nutrients. Building on conceptual and mathematical models developed for highly eutrophic river systems in Europe and North America (e.g., Garnier and Billen, 2007), we need to develop new integrative frameworks to explain river-specific responses to the unprecedented pace and scale of urbanization and water pollution. These integrative frameworks need to consider concurrent multiple environmental changes including dams and climatic variability and extreme events as confounding factors that can either boost or dampen pollution-induced pulses of CO₂ emissions from
- 530 highly polluted urban rivers. A key future challenge in predicting CO_2 emissions from highly polluted, eutrophic river systems would be to constrain shifting balances between interrelated riverine metabolic processes, as illustrated by the regime shift in the relationship between Chl *a* and *p*CO₂ observed between the less impacted upstream and eutrophic downstream reaches of a highly urbanized river system (Fig. 5).

How to overcome the overall scarcity and spatially uneven availability of high-quality data, guided by an integrative
conceptual framework reflecting observed regional trends, might be the number one research priority in providing a scientifically robust assessment of the current status of human impacts on C fluxes of Asian river systems. Given the inadequate research capacity in many developing Asian countries, more efforts should be given to build collaborative research networks that can provide researchers with practical guides and standardized methodologies for designing and conducting field monitoring of riverine C fluxes at multiple spatial and temporal scales. These efforts need to pay more attention to emerging local issues in addition to the common regional patterns associated with river impoundment and pollution. As observed in the Tibetan Plateau, urbanization and dam construction have been expanding to upstream

- pollution. As observed in the Tibetan Plateau, urbanization and dam construction have been expanding to upstream headwater reaches of the large rivers such as the Mekong, Yangtze, and Yellow River. The rapid expansion of anthropogenic perturbations, coupled with idiosyncratic local climates and ecosystems, can amplify changes in riverine metabolic processes and C dynamics. To better assess the interactive effects of concurrent multiple environmental changes and human-induced
- 545 perturbations to the riverine networks of interacting land and water patches, we suggest that the long-standing concept of river continuum assuming 'natural' states should be critically examined by field measurements and complemented with alternative perspectives of 'discontinuity' or 'discontinuous continuity' (*sensu* Poole, 2002) in riverine metabolisms and C fluxes created in impounded and eutrophic reaches of rivers draining increasingly urbanizing watersheds across Asia and globally.

550 Data availability

Data are available and can be requested from the corresponding author (jhp@ewha.ac.kr).

Author contribution

All authors contributed to data acquisition, the discussion of concepts and research topics, and manuscript preparation. The manuscript was written through concerted efforts of all authors, coordinated by J.-H. Park.

555 Competing interests

The authors declare that they have no conflict of interest.

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References

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- Abril, G., Guérin, F., Richard, S., Delmas, R., Galy-Lacaux, C., Gosse, P., Tremblay, A., Varfalvy, L., Dos Santos, M. A., and Matvienko, B.: Carbon dioxide and methane emissions and the carbon budget of a 10-year old tropical reservoir (Petit Saut, French Guiana), Global Biogeochem. Cycles, 19, GB4007, doi:10.1029/2005GB002457, 2005.
- 565 Abril, G., Martinez, J.-M., Artigas L. F., Moreira-Turcq P., Benedetti M. F., Vidal L., Meziane T., Kim J.-H., Bernardes M. C., Savoye N., Deborde J., Albéric P., Souza M. F. L., Souza E. L., and Roland F.: Amazon River carbon dioxide outgassing fuelled by wetlands, Nature, 505, 395–398, doi:10.1038/nature12797, 2014.
 - Abril, G., Bouillon, S., Darchambeau, F., Teodoru, C. R., Marwick, T. R., Tamooh, F., Ochieng Omengo, F., Geeraert, N., Deirmendjian, L., Polsenaere, P., and Borges, A.V.: Technical Note: Large overestimation of pCO₂ calculated from pH
- 570 and alkalinity in acidic, organic-rich freshwaters. Biogeosciences, 12, 67-78, doi:10.5194/bg-12-67-2015, 2015.
 - Alin, S. R., Rasera, M. F. F. L., Salimon, C. I., Richey, J. E., Holtgrieve, G. W., Krusche, A. V., and Snidvongs, A.: Physical controls on carbon dioxide transfer velocity and flux in low-gradient river systems and implications for regional carbon budgets, J. Geophys. Res., 116, G01009. doi:10.1029/2010JG001398, 2011.

Allan, J. D.: Landscapes and riverscapes: The influence of land use on stream ecosystems, Annu. Rev. Ecol. Evol. Syst, 35,

- 575 257–284, doi:10.1146/annurev.ecolsys.35.120202.110122, 2004.
 - Amann, T., Weiss, A., and Hartmann, J.: Carbon dynamics in the freshwater part of the Elbe estuary, Germany: Implications of improving water quality, Estuar. Coast. Shelf Sci., 107, 112–121, doi:10.1016/j.ecss.2012.05.012, 2012.
 - Barros, N., Cole, J. J., Tranvik, L. J., Prairie, Y. T., Bastviken, D., Huszar, V. L. M., del Giorgio, P. and Roland, F.: Carbon emission from hydroelectric reservoirs linked to reservoir age and latitude, Nat. Geosci., 4, 593–596, doi:10.1038/ngeo1211, 2011.

Battin, T. J., Luyssaert, S., Kaplan, L.A., Aufdenkampe, A. K., Richter, A., and Tranvik, L. J.: The boundless carbon cycle, Nat. Geosci., 2, 598–600, doi:10.1038/ngeo618, 2009. Baum, A., Rixen, T., and Samiaji, J.: Relevance of peat draining rivers in central Sumatra for the riverine input of dissolved organic carbon into the ocean. Estuar. Coast. Shelf Sci., 73, 563–570, doi:10.1016/j.ecss.2007.02.012, 2007.

- Bhatt, M. P., McDowell, W. H., Gardner, K. H., and Hartmann, J.: Chemistry of the heavily urbanized Bagmati River system 585 in Kathmandu Valley, Nepal: export of organic matter, nutrients, major ions, silica, and metals, Environ. Earth Sci., 71, 911-922, doi:10.1007/s12665-013-2494-9, 2014.
 - Bickle, M. J., Bunbury, J., Chapman, H. J., Harris, N. B. W., Fairchild, I. J., and Ahmed, T.: Fluxes of Sr into the headwaters of the Ganges, Geochim. Cosmochim. Acta, 67, 2567–2584, doi:10.1016/S0016-7037(03)00029-2, 2003.
- Biswas, H., Mukhopadhyay, S. K., De, T. K., Sen, S., and Jana, T. K.: Biogenic controls on the air-water carbon dioxide 590 exchange in the Sundarban mangrove environment, northeast coast of Bay of Bengal, India, Limnol. Oceanogr., 49, 95– 101, doi:10.4319/lo.2004.49.1.0095, 2004.
 - Borges, A. V., and Abril, G.: Carbon dioxide and methane dynamics in estuaries, in: Treatise on Estuarine and Coastal Science, Volume 5, Wolanski, E., and McLusky, D. (Eds.), Academic Press, Waltham, 119-161, doi:10.1016/B978-0-

595 12-374711-2.00504-0, 2011.

- Borges, A. V., Schiettecatte, L.-S., Abril, G., Delille, B., and Gazeau, F.: Carbon dioxide in European coastal waters, Estuar. Coast. Shelf Sci., 70, 375-387, doi:10.1016/j.ecss.2006.05.046, 2006.
- Borges, A. V., Abril, G., and Bouillon, S.: Carbon dynamics in the Mekong Delta. Biogeosciences Discuss., doi.org/10.5194/bg-2017-444, 2017, In review.
- 600 Butman, D. E., Wilson, H. F., Barnes, R. T., Xenopoulos, M. A., and Raymond, P. A.: Increased mobilization of aged carbon to rivers by human disturbance, Nat. Geosci., 8, 112–116, doi:10.1038/ngeo2322, 2015.
 - Cadenasso, M. L., Pickett, S. T., Groffman, P. M., Band, L. E., Brush, G. S., Galvin, M. F., Grove, J. M., Hagar, G., Marshall, V., McGrath, B. P., O'Neil-Dunne, J. P., Stack, W. P., and Troy, A. R.: Exchanges across land-water-scape boundaries in urban systems: strategies for reducing nitrate pollution, Ann. N. Y. Acad. Sci., 1134, 213-232,
- 605 doi:10.1196/annals.1439.012, 2008.
 - Cai, W. J., Guo, X. H., Chen, C.-T. A., Dai, M. H., Zhang, L. J., Zhai, W. D., Lohrenz, S. E., Yin, K., Harrison, P. J., and Wang, Y. C.: A comparative overview of weathering intensity and HCO₃ flux in the world's largest rivers with

emphasis on the Changjiang, Huanghe, Zhujiang (Pearl) and Mississippi Rivers, Continental Shelf Research, 28, 1538– 1549, doi:10.1016/j.csr.2007.10.014, 2008.

- 610 Casas-Ruiz, J. P., Tittel, J., von Schiller, D., Catalán, N., Obrador, B., Gómez-Gener, L., Zwirnmann, E., Sabater, S., and Marce, R.: Drought-induced discontinuities in the source and degradation of dissolved organic matter in a Mediterranean river, Biogeochemistry, 127, 125–139, doi:10.1007/s10533-015-0173-5, 2016.
 - Catalán, N., Marcé R., Kothawala, D. N., and Tranvik, L. J.: Organic carbon decomposition rates controlled by water retention time across inland waters, Nat. Geosci., 9, 501–504, doi:10.1038/ngeo2720, 2016.
- 615 Chakrapani, G. J., and Veizer, J.: Dissolved inorganic carbon isotopic compositions in the Upstream Ganga river in the Himalayas, Curr. Sci., 89, 553–556, 2005.
 - Chen, H., Wu, Y., Yuan, X., Gao, Y., Wu, N., and Zhu, D.: Methane emissions from newly created marshes in the drawdown area of the Three Gorges Reservoir, J. Geophys. Res., 114, D18301, doi:10.1029/2009JD012410, 2009.

- 620 doi:10.1038/ngeo2544, 2015.
 - Chou, W.-C., Gong, G.-C., Cai, W.-J., and Tseng, C.-M.: Seasonality of CO₂ in coastal oceans altered by increasing anthropogenic nutrient delivery from large rivers: evidence from the Changjiang–East China Sea system, Biogeosciences, 10, 3889–3899, doi:10.5194/bg-10-3889-2013, 2013.
 - Cole, J. J., Prairie, Y. T., Caraco, N. F., McDowell, W. H., Tranvik, L. J., Striegl, R. G., Duarte, C. M., Kortelainen, P.,
- Downing, J. A., Middelburg, J. J., and Melack, J.: Plumbing the global carbon cycle: Integrating inland waters into the terrestrial carbon budget, Ecosystems, 10, 171–184, doi:10.1007/s10021-006-9013-8, 2007.
 - Crawford, J. T., Loken, L. C., Stanley, E. H., Stets, E. G., Dornblaser, M. M., and Striegl, R. G.: Basin scale controls on CO₂ and CH₄ emissions from the Upper Mississippi River, Geophys. Res. Lett. 43, 1973–1979. doi:10.1002/2015GL067599, 2016.
- 630 Dai, M., Yin, Z., Meng, F., Liu, Q., and Cai, W.-J.: Spatial distribution of riverine DOC inputs to the ocean: an updated global synthesis, Curr. Opin. Environ. Sustain., 4, 170–178, doi:10.1016/j.cosust.2012.03.003, 2012.

Das, A. K.: Limno-chemistry of some Andhra Pradesh reservoirs, J. Inland Fish. Soc. India, 32, 37–44, 2000.

Chen, Y., Wang, K., Lin, Y., Shi, W., Song, Y., and He, X.: Balancing green and grain trade, Nat. Geosci., 8, 739-741,

- Degens, E. T., Kempe, S., and Richey, J. E.: Summary: biogeochemistry of major world river, in: Biogeochemistry of Major World Rivers, SCOPE Volume 42, Degens, E. T., Kempe, S., and Richey, J. E. (eds.), 323-347, 1991.
- 635 Deshmukh, C., Serça, D., Delon, C., Tardif, R., Demarty, M., Jarnot, C., Meyerfeld, Y., Chanudet, V., Guédant, P., Rode, W., Descloux, S., and Guérin, F.: Physical controls on CH₄ emissions from a newly flooded subtropical freshwater hydroelectric reservoir: Nam Theun 2, Biogeosciences, 11, 4251–4269, doi:10.5194/bg-11-4251-2014, 2016.
 - Deshmukh, C., Guérin, F., Vongkhamsao, A., Pighini, S., Oudone, P., Sopraseuth, S., Godon, A., Rode, W., Guédant, P., Oliva, P., Audry, S., Zouiten, C., Galy-Lacaux, C., Robain, H., Ribolzi, O., Kansal, A., Chanudet, V., Descloux, S., and
- 640 Serça, D.: Carbon dioxide emissions from the flat bottom and shallow Nam Theun 2 Reservoir: drawdown area as a neglected pathway to the atmosphere, Biogeosciences, doi:10.5194/bg-2017-380, 2017, In review.
 - Dutta, M. K., Mukherjee, R., Jana, T. K., and Mukhopadhyay, S. K.: Biogeochemical dynamics of exogenous methane in an estuary associated to a mangrove biosphere; the Sundarbans, NE coast of India, Mar. Chem., 170, 1–10, doi:10.1016/j.marchem.2014.12.006, 2015.
- 645 Ellis, E. E., Keil, R. G., Ingalls, A. E., and Richey, J. E.: Seasonal variability in the sources of particulate organic matter of the Mekong River as discerned by elemental and lignin analyses, J. Geophys. Res., 117, G01038, doi:10.1029/2011JG001816, 2012.
 - Evans, A. E. V., Hanjra, M. A., Jiang, Y., Qadir, M., and Drechsel, P.: Water quality: Assessment of the current situation in Asia, Int. J. Water Resour. Dev., 28, 195–216, doi:10.1080/07900627.2012.669520, 2012.
- 650 FAO (Food and Agriculture Organization), AQUASTAT (http://www.fao.org/nr/water/aquastat/sets/index.stm, retrieved on 15. 12. 2017)
 - Feng, X., Fu, B., Lu, N., Zeng, Y., and Wu, B.: How ecological restoration alters ecosystem services: an analysis of carbon sequestration in China's Loess Plateau, Sci. Rep., 3, 2846, doi:10.1038/srep02846, 2013.

Frankignoulle, M., Abril, G., Borges, A., Bourge, I., Canon, C., Delille, B., Libert, E., and Théate, J.-M.: Carbon dioxide emission from European estuaries, Science, 282, 434–436, doi:10.1126/science.282.5388.434, 1998.

Galy, V., Peucker-Ehrenbrink, B., and Eglinton, T.: Global carbon export from the terrestrial biosphere controlled by erosion, Nature, 521, 204–207, doi:10.1038/nature14400, 2015.

Garnier, J., and Billen, G.: Autotrophy and heterotrophy of aquatic communities in the Seine river system, Sci. Total Environ., 375, 110–124, 2007.

- 660 Griffith, D. R., Barnes, R. T., and Raymond, P. A.: Inputs of fossil carbon from wastewater treatment plants to U.S. rivers and oceans, Environ. Sci. Technol., 43, 5647–5651, doi:10.1021/es9004043, 2009.
 - Griffith, D. R., and Raymond, P. A.: Multiple-source heterotrophy fueled by aged organic carbon in an urbanized estuary, Mar. Chem., 124, 14–22, doi:10.1016/j.marchem.2010.11.003, 2011.
 - Grimm, N. B., Faeth, S. H., Golubiewski, N. E., Redman, C. L., Wu, J., Bai, X., and Briggs, J. M.: Global change and the ecology of cities, Science, 319, 756–760, doi:10.1126/science.1150195, 2008.
 - Grumbine, R. E., Dore, J., and Xu, J.: Mekong hydropower: drivers of change and governance challenges, Front. Ecol. Environ., 10, 91–98, doi:10.1890/110146, 2012.
 - Guo, W., Yang, L., Zhai, W., Chen, W., Osburn, C. L., Huang, X., and Li, Y.: Runoff-mediated seasonal oscillation in the dynamics of dissolved organic matter in different branches of a large bifurcated estuary—The Changjiang Estuary, J.
- 670 Geophys. Res. Biogeosci., 119, 776–793, doi:10.1002/2013JG002540, 2014.

665

- Gupta, G. V. M., Thottathil, S. D., Balachandran, K. K., Madhu, N. V., Madeswaran, P., and Nair, S.: CO₂ supersaturation and net heterotrophy in a tropical estuary (Cochin, India): Influence of anthropogenic effect, Ecosystems, 12, 1145– 1157, doi:10.1007/s10021-009-9280-2, 2009.
- Hartmann, J., Jansen, N., Kempe, S., and H. Dürr, H., Geochemistry of the river Rhine and the upper Danube: Recent trends and lithological influence on baselines, J. Environ. Sci. Sustain. Soc., 1, 39–46, 2007.
- Hartmann, J., Lauerwald, R., and Moosdorf, N., A Brief Overview of the GLObal RIver Chemistry Database, GLORICH. Procedia Earth Planet. Sci., 10, 23–27, doi:10.1016/j.proeps.2014.08.005, 2014.
- Hilton, J., O'Hare, M., Bowes, M. J., and Jones, J. I.: How green is my river? A new paradigm of eutrophication in rivers, Sci. Total Environ., 365, 66–83, doi:10.1016/j.scitotenv.2006.02.055, 2006.
- 680 Hosen, J. D., McDonough, O. T., Febria, C. M., and Palmer, M. A.: Dissolved organic matter quality and bioavailability changes across an urbanization gradient in headwater streams, Environ. Sci. Technol., 48, 7817–7824, doi:10.1021/es501422z, 2014.

Hotchkiss, E. R., Hall Jr, R. O., Sponseller, R. A., Butman, D., Klaminder, J., Laudon, H., Rosvall, M., and Karlsson, J.: Sources of and processes controlling CO₂ emissions change with the size of streams and rivers, Nat. Geosci., 8, 696–

- 685 699, doi:10.1038/ngeo2507, 2015.
 - Hu, Y., and Cheng, H.: The urgency of assessing the greenhouse gas budgets of hydroelectric reservoirs in China, Nat. Clim. Change, 3, 708–712, doi:10.1038/nclimate1831, 2013.
 - Huang, X., Sillanpaa, M., Gjessing, E. T., Peraniemi, S., and Vogt, R. D.: Water quality in the southern Tibetan Plateau: chemical evaluation of the Yarlung Tsangpo (Brahmaputra), River Res. Applic., 27, 113–121, doi:10.1002/rra.1332,

690

2011.

- Huang, T.-H., Fu, Y.-H., Pan, P.-Y., and Chen, C.-T. A.: Fluvial carbon fluxes in tropical rivers, Curr. Opin. Environ. Sustain., 4, 162–169, doi:10.1016/j.cosust.2012.02.004, 2012.
- Hynes, H. B. N.: The stream and its valley, Verh. Int. Ver. Theor. Angew. Limnol., 19, 1–15, doi:0.1080/03680770.1974.11896033, 1975.
- 695 Irvine, K. N, Murphy, T., Sampson, M., Dany, V., Vermette, S. J., and Tang, T.: An overview of water quality issues in Cambodia, Journal of Water Management Modeling, R225-02, doi:10.14796/JWMM.R225-02, 2006.
 - Ittekkot, V., Safiullah, S., Mycke, B., and Seifert, R.: Seasonal variability and geochemical significance of organic matter in the River Ganges, Bangladesh, Nature, 317, 800–802, doi:10.1038/317800a0, 1985.

Ittekkot, V.: Global trends in the nature of organic matter in river suspensions, Nature, 332, 436–438, doi:10.1038/332436a0,

700

1988.

Jin, H., Yoon, T. K., Lee, S.-H., Kang, H., Im, J., and Park, J.-H.: Enhanced greenhouse gas emission from exposed sediments along a hydroelectric reservoir during an extreme drought event, Environ. Res. Lett., 11, 124003, doi:10.1088/1748-9326/11/12/124003, 2016.

Joesoef, A., Huang, W.-J., Gao, Y., and Cai, W.-J.: Air–water fluxes and sources of carbon dioxide in the Delaware Estuary: spatial and seasonal variability, Biogeosciences, 12, 6085–6101, doi:10.5194/bg-12-6085-2015, 2015.

- Jung, B.-J., Lee, H.-J., Jeong, J.-J., Owen, J., Kim, B., Meusburger, K., Alewell, C., Gebauer, G., Shope, C., and Park, J.-H.: Storm pulses and varying sources of hydrologic carbon export from a mountainous watershed, J. Hydrol., 440–441, 90-101, doi:10.1016/j.jhydrol.2012.03.030, 2012.
- Kaushal, S. S., and Belt, K. T.: The urban watershed continuum: evolving spatial and temporal dimensions, Urban Ecosyst., 15, 409–435, doi:10.1007/s11252-012-0226-7, 2012.
- Kaushal, S. S., McDowell, W. H., and Wollheim, W. M.: Tracking evolution of urban biogeochemical cycles: past, present, and future, Biogeochemistry, 121, 1–21, doi:10.1007/s10533-014-0014-y, 2014.
- Kempe, S.: Long-term records of CO₂ pressure fluctuations in fresh water, in Transport of Carbon and Minerals in Major World Rivers Part 1, Degens, E. T. (Ed.), SCOPE/UNEP Sonderband 52, Mitt. Geol.-Paläont. Inst. Univ. Hamburg,

715 Hamburg, 91–332, 1982.

710

- Kempe, S.: Sinks of the anthropogenically enhanced carbon-cycle in surface fresh waters, J. Geophys. Res., 89, 4657–4676, doi:10.1029/JD089iD03p04657, 1984.
- Koehler, B., von Wachenfeldt, E., Kothawala, D., and Tranvik, L. J.: Reactivity continuum of dissolved organic carbon decomposition in lake water, J. Geophys. Res., 117, G01024, doi:10.1029/2011JG001793, 2012.
- 720 Krishna, M. S., Prasad, M. H. K., Rao, D. B., Viswanadham, R., Sarma, V. V. S. S., and Reddy, N. P. C.: Export of dissolved inorganic nutrients to the northern Indian Ocean from the Indian monsoonal rivers during discharge period, Geochim. Cosmochim. Acta, 172, 430–443, doi:10.1016/j.gca.2015.10.013, 2015.
 - Labat, D., Goddéris, Y., Probst, J. L., and Guyot, J. L.: Evidence for global runoff increase related to climate warming, Adv. Water Resour., 27, 631–642, doi:10.1016/j.advwatres.2004.02.020, 2004.
- 725 Lauerwald, R., Laruelle, G. G., Hartmann, J., Ciais, P., and Regnier, P. A. G.: Spatial patterns in CO₂ evasion from the global river network, Global Biogeochem. Cycles, 29, 534–554, doi:10.1002/2014GB004941, 2015.
 - Lehner, B., Liermann, C. R., Revenga, C., Vörösmarty, C., Fekete, B., Crouzet, P., Döll, P., Endejan, M., Frenken, K., Magome, J., Nilsson, C., Robertson, J. C., Rödel, R., Sindorf, N., and Wisser, D.: High-resolution mapping of the world's reservoirs and dams for sustainable river-flow management, Front. Ecol. Environ., 9, 494–502, doi:10.1890/100125, 2011.

- Le, T.P.Q., Marchand, C., Ho, C.T., Duong, T.T., Nguyen, H.T.M., XiXi, L., Vu, D.A., Doan, P.K., and Le, N.D.: CO₂ partial pressure and CO₂ emissions from the lower Red River (Vietnam), Biogeosciences Discuss., https://doi.org/10.5194/bg-2017-505, 2017.
- Li, S., and Bush, R. T.: Changing fluxes of carbon and other solutes from the Mekong River, Sci. Rep., 5, 16005,
- 735 doi:10.1038/srep16005, 2015.

- Li, G., Xia, X., Yang, Z., Wang, R., and Voulvoulis, N.: Distribution and sources of polycyclic aromatic hydrocarbons in the middle and lower reaches of the Yellow River, China, Environ. Pollut., 144, 985–993, doi:10.1016/j.envpol.2006.01.047, 2006.
- Li, S.Y., Lu, X.X., He, M., Zhou, Y., Li, L., Ziegler, A.D.: Daily CO₂ partial pressure and CO₂ outgassing in the upper
- 740 Yangtze River basin: a case study of the Longchuan River, China. J. Hydrol., 466–467, 141–150, doi.org/10.1016/j.jhydrol.2012.08.011, 2012.
 - Li, J., Dong, S., Liu, S., Yang, Z., Peng, M., and Zhao, C.: Effects of cascading hydropower dams on the composition, biomass and biological integrity of phytoplankton assemblages in the middle Lancang-Mekong River, Ecol. Eng., 60, 316–324, doi:10.1016/j.ecoleng.2013.07.029, 2013a.
- Li, S., Lu, X. X., and Bush, R. T.: CO₂ partial pressure and CO₂ emission in the Lower Mekong River, J. Hydrol., 504, 40–56, doi:10.1016/j.jhydrol.2013.09.024, 2013b.
 - Liu, S., Lu, X. X., Xia, X., Zhang, S., Ran, L., Yang, X., and Liu, T.: Dynamic biogeochemical controls on river pCO₂ and recent changes under aggravating river impoundment: An example of the subtropical Yangtze River, Global Biogeochem. Cycles, 30, 880–897, doi:10.1002/2016GB005388, 2016.
- Lookingbill, T. R., Kaushal, S. S., Elmore, A. J., Gardner, R., Eshleman, K. N., Hilderbrand, R. H., Morgan, R. P., Boynton,
 W. R., Palmer, M. A., and Dennison, W. C.: Altered ecological flows blur boundaries in urbanizing watersheds, Ecol.
 Soc. 14, 10, doi:10.5751/ES-02989-140210, 2009.
 - Lu, Y., Song, S., Wang, R., Liu, Z., Meng, J., Sweetman, A. J., Jenkins, A., Ferrier, R. C., Li, H., and Luo, W.: Impacts of soil and water pollution on food safety and health risks in China, Environ. Int., 77, 5–15, doi:10.1016/j.envint.2014.12.010, 2015.

- Ludwig, W., Probst, J.-L., and Kempe, S.: Predicting the oceanic input of organic carbon by continental erosion, Global Biogeochem. Cycles, 10, 23–41, doi: 10.1029/95GB02925, 1996.
- Luthy, R. G., Sedlak, D.L., Plumlee, M. H., Austin, D., and Resh, V. H.: Wastewater-effluent-dominated streams as ecosystem-management tools in a drier climate, Front. Ecol. Environ., 13, 477–485, doi:10.1890/150038, 2015.
- 760 Maavara, T., Lauerwald, R., Regnier, P., and Van Cappellen, P.: Global perturbation of organic carbon cycling by river damming, Nat. Commun., 15347, doi:10.1038/ncomms15347, 2017.
 - Manaka, T., Otani, S., Inamura, A., Suzuki, A., Aung, T., Roachanakanan, R., Ishiwa, T., and Kawahata, H.: Chemical weathering and long-term CO₂ consumption in the Ayeyarwady and Mekong river basins in the Himalayas, J. Geophys. Res. Biogeosci., 120, 1165–1175, doi:10.1002/2015JG002932, 2015a.
- 765 Manaka, T., Ushie, H., Araoka, D., Otani, S., Inamura, A., Suzuki, A., Zakir Hossain, H. M., and Kawahata, H.: Spatial and Seasonal Variation in Surface Water pCO₂ in the Ganges, Brahmaputra, and Meghna Rivers on the Indian Subcontinent, Aquat. Geochem., 21, 437-458, doi:10.1007/s10498-015-9262-2, 2015b.
 - Martin, E. E., Ingalls, A. E., Richey, J. E., Keil, R. G., Santos, G. M., Truxal, L. T., Alin, S. R., and Druffel, E. R. M.: Age of riverine carbon suggests rapid export of terrestrial primary production in tropics, Geophys. Res. Lett., 40, 5687–5691,

770 doi:10.1002/2013GL057450, 2013.

Marx, A., Dusek, J., Jankovec, J., Sanda, M., Vogel, T., van Geldern, R., Hartmann, J., and Barth, J. A. C.: A review of CO₂ and associated carbon dynamics in headwater streams: A global perspective, Rev. Geophys., 55, 560–585, doi:10.1002/2016RG000547, 2017.

Mateo-Sagasta, J., Raschid-Sally, L., and Thebo, A.: Global wastewater and sludge production, treatment and use, in:

- Wastewater–Economic Asset in an Urbanizing World. Drechsel, P., Qadir, M., Wichelns, D. (Eds), Springer, Dordrecht,
 15–38, doi:10.1007/978-94-017-9545-6_2, 2015.
 - McCluney, K. E., Poff, N. L., Palmer, M. A., Thorp, J. H., Poole, G. C., Williams, B. S., Williams, M. R., and Baron, J. S.: Riverine macrosystems ecology: sensitivity, resistance, and resilience of whole river basins with human alterations, Front. Ecol. Environ., 12, 48–5, doi:10.1890/120367, 2014.

- Meybeck, M., and Helmer, R.: The quality of rivers: From pristine stage to global pollution, Glob. Planet. Change, 1, 283–309, doi:10.1016/0921-8181(89)90007-6, 1989.
 - Milliman, J. D., Farnsworth, K. L.: River Discharge to the Coastal Ocean: A Global Synthesis, Cambridge University Press, Cambridge, UK, 2011.
 - Milliman, J. D., Farnsworth, K. L., Jones, P. D., Xu, K. H., and Smith, L. C.: Climatic and anthropogenic factors affecting
- 785 river discharge to the global ocean, 1951–2000, Global Planet. Change, 62, 187–194, doi:10.1016/j.gloplacha.2008.03.001, 2008.
 - Milly, P. C. D., Dunne, K. A., and Vecchia, A. V.: Global pattern of trends in streamflow and water availability in a changing climate, Nature, 438, 347–350, doi:10.1038/nature04312, 2005.

Min, S.-K., Zhang, X., Zwiers, F. W., and Hegerl, G. C.: Human contribution to more-intense precipitation extremes, Nature,

790

0 470, 378–381, doi:10.1038/nature09763, 2011.

- Mosher, J. J., Kaplan, L. A., Podgorski, D. C., McKenna, A. M., and Marshall, A. G.: Longitudinal shifts in dissolved organic matter chemogeography and chemodiversity within headwater streams: a river continuum reprise, Biogeochemistry, 124, 371-385, doi.org/10.1007/s10533-015-0103-6, 2015.
- Mukhopadhyay, S. K., Biswas, H., De, T. K., Sen, S., and Jana, T. K.: Seasonal effects on the air-water carbon dioxide
- 795 exchange in the Hooghly estuary, NE coast of Bay of Bengal, India, J. Environ. Monit., 4, 549–552, doi:10.1039/B201614A, 2002.
 - Palmer, M. A., Liermann, C. A. R., Nilsson, C., Flörke, M., Alcamo, J., Lake, P.S., and Bond, N.: Climate change and the world's river basins: anticipating management options, Front. Ecol. Environ., 6, 81–89, doi:10.1890/060148, 2008.
 - Panneer Selvam, B., Natchimuthu, S., Arunachalam, L., and Bastviken, D.: Methane and carbon dioxide emissions from
- inland waters in India implications for large scale greenhouse gas balances, Glob. Change Biol., 20, 3397–3407,
 doi:10.1111/gcb.12575, 2014.
 - Park, J.-H., Duan, L., Kim, B., Mitchell, M. J., and Shibata, H.: Potential effects of climate change and variability on watershed biogeochemical processes and water quality in Northeast Asia, Environ. Int., 36, 212–225, doi:10.1016/j.envint.2009.10.008, 2010.

- 805 Park, J.-H., Inam, E., Abdullah, M. H., Agustiyani, D., Duan, L., Hoang, T. T., Kim, K.-W., Kim, S. D., Nguyen, M. H., Pekthong, T., Sao, V., Sarjiya, A., Savathvong, S., Sthiannopkao, S., Syers, J. K., and Wirojanagud, W.: Implications of rainfall variability for seasonality and climate-induced risks concerning surface water quality in East Asia, J. Hydrol., 400, 323–332, doi:10.1016/j.jhydrol.2011.01.050, 2011.
- Paul, M. J., and Meyer, J. L.: Streams in the urban landscape, Annu. Rev. Ecol. Syst., 32, 333–365, doi:10.1146/annurev.ecolsys.32.081501.114040, 2001.
 - Pierrot, D., Lewis, E., and Wallace, D. W. R.: MS Excel Program developed for CO₂ system calculations, ORNL/CDIAC-105a, Carbon dioxide Information Analysis Center, Oak Ridge National Laboratory, U.S. Department of Energy, Oak Ridge, Tennessee, doi:10.3334/CDIAC/otg.CO2SYS_XLS_CDIAC105a, 2006.

Poole, G. C.: Fluvial landscape ecology: addressing uniqueness within the river discontinuum, Freshwater Biol., 47, 641–660,

815 doi:10.1046/j.1365-2427.2002.00922.x, 2002.

820

- Poole, G. C.: Stream hydrogeomorphology as a physical science basis for advances in stream ecology, J. North Am. Benthol. Soc., 29, 12–25, doi:10.1899/08-070.1, 2010.
- Pradhan, U. K., Wu, Y., Shirodkar, P. V., Zhang, J., and Zhang, G.: Multi-proxy evidence for compositional change of organic matter in the largest tropical (peninsular) river basin of India, J. Hydrol., 519, 999–1009, doi:10.1016/j.jhydrol.2014.08.018, 2014.
- Prasad, M. H. K., Sarma, V. V. S. S., Sarma, V. V., Krishna, M. S., and Reddy, N. P. C.: Carbon Dioxide Emissions from the Tropical Dowleiswaram Reservoir on the Godavari River, Southeast of India, Journal of Water Resource and Protection, 5, 534–545, doi:10.4236/jwarp.2013, 2013.
 - Qu, B., Sillanpaa, M., Zhang, Y., Guo, J., Wahed, M. S. M. A., and Kang, S.: Water chemistry of the headwaters of the Yangtze River, Environ. Earth Sci., 74, 6443–6458, doi:10.1007/s12665-015-4174-4, 2015.
- Qu, B., Zhang, Y., Kang, S., and Sillanpaa, M.: Water chemistry of the southern Tibetan Plateau: an assessment of the Yarlung Tsangpo river basin, Environ. Earth Sci., 76, 74, doi:10.1007/s12665-017-6393-3, 2017.
- Ramesh, R., and Subramanian, V.: Temporal, spatial and size variation in sediment transport in the Krishna River Basin, India, J. Hydrol., 98, 53–65, doi:10.1016/0022-1694(88)90205-3, 1988.

- 830 Ramesh, R., Robin, R. S., and Purvaja, R.: An inventory on the phosphorus flux of major Indian rivers, Curr. Sci., 108, 1294–1299, 2015.
 - Ran, L., Lu, X. X., Sun, H., Han, J., Li, R., and Zhang, J.: Spatial and seasonal variability of organic carbon transport in the Yellow River, China, J. Hydrol., 498, 76–88, doi:10.1016/j.jhydrol.2013.06.018, 2013a.
 - Ran, L., Lu, X. X., Xin, Z., and Yang, X.: Cumulative sediment trapping by reservoirs in large river basins: A case study of
- the Yellow River basin, Global Planet. Change, 100, 308–319, doi:10.1016/j.gloplacha.2012.11.001, 2013b.
 - Ran, L., Lu, X. X., and Xin, Z.: Erosion-induced massive organic carbon burial and carbon emission in the Yellow River basin China, Biogeosciences, 11, 945–959, doi:10.5194/bg-11-945-2014, 2014.
 - Ran, L., Lu, X. X., Richey, J. E., Sun, H., Han, J., Liao, S., and Yi, Q.: Long-term spatial and temporal variation of CO₂ partial pressure in the Yellow River, China, Biogeosciences, 12, 921-932, doi:10.5194/bg-12-921-2015, 2015a.
- Ran, L., Lu, X. X., Yang, H., Li, L., Yu, R., Sun, H., and Han, J.: CO₂ outgassing from the Yellow River network and its implications for riverine carbon cycle, J. Geophys. Res. Biogeosci., 120, 1334–1347, doi:10.1002/2015JG002982, 2015b.
 - Ran, L., Li, L., Tian, M., Yang, X., Yu, R., Zhao, J., Wang, L., and Lu, X. X.: Riverine CO₂ emissions in the Wuding River catchment on the Loess Plateau: Environmental controls and dam impoundment impact, J. Geophys. Res. Biogeosci.,

845 122, 1439–1455, doi:10.1002/2016JG003713, 2017a.

- Ran, L., Lu, X. X., and Liu, S.: Dynamics of riverine CO₂ in the Yangtze River fluvial network and their implications for carbon evasion, Biogeosciences, 14, 2183–2198, doi:10.5194/bg-14-2183-2017, 2017b.
- Raymond, P. A., and Spencer, R. G. M.: Riverine DOM, in: Biogeochemistry of Marine Dissolved Organic Matter, Second Edition, Hansell, D. A. and Carlson, C. A. (Eds.), Academic Press, Boston, 509–533, doi:10.1016/B978-0-12-405940-5.00011-X, 2015.
- Raymond, P. A., Hartmann, J., Lauerwald, R., Sobek, S., McDonald, C., Hoover, M., Butman, D., Striegl, R., Mayorga, E., Humborg, C., Kortelainen, P., Dürr, H., Meybeck, M., Ciais, P., and Guth, P.: Global carbon dioxide emissions from inland waters, Nature, 503, 355–359, doi:10.1038/nature12760, 2013.

Regnier, P., Friedlingstein, P., Ciais, P., Mackenzie, F. T., Gruber, N., Janssens, I. A., Laruelle, G. G., Lauerwald, R.,

- Luyssaert, S., Andersson, A. J., Arndt, S., Arnosti, C., Borges, A. V., Dale, A. W., Gallego-Sala, A., Goddéris, Y.,
 Goossens, N., Hartmann, J., Heinze, C., Ilyina, T., Joos, F., LaRowe, D. E., Leifeld, J., Meysman, F. J. R., Munhoven,
 G., Raymond, P. A., Spahni, R., Suntharalingam, P., and Thullner, M.: Anthropogenic perturbation of the carbon fluxes
 from land to ocean, Nat. Geosci., 6, 597–607, doi:10.1038/ngeo1830, 2013.
 - Samanta, S., Dalai, T. K., Pattanaik, J. K., Rai, S. K., and Mazumdar, A.: Dissolved inorganic carbon (DIC) and its δ^{13} C in
- 860 the Ganga (Hooghly) River estuary, India: Evidence of DIC generation via organic carbon degradation and carbonate dissolution, Geochim. Cosmochim. Acta, 165, 226–248, doi:10.1016/j.gca.2015.05.040, 2015.
 - Sarin, M. M., Krishnaswaswami, S., Dilli, K., Somayajulu, B. L. K., and Moore W. S.: Major ion chemistry of the Ganga-Brahmaputra river system: Weathering processes and fluxes to the Bay of Bengal, Geochim. Cosmochim. Acta, 53, 997–1009, doi:10.1016/0016-7037(89)90205-6, 1989.
- 865 Sarma, V. V. S. S., Gupta, N. M., Babu, P. V. R., Acharya, T., Harikrishnachari, N., Vishnuvardhan, K., Rao, N. S., Reddy, N. P. C., Sarma, V. V., Sadhuram, Y., Murty, T. V. R., and Kumar, M. D.: Influence of river discharge on plankton metabolic rates in the tropical monsoon driven Godavari estuary, India, Estuar. Coast. Shelf Sci., 85, 515–524, doi:10.1016/j.ecss.2009.09.003, 2009.
 - Sarma, V. V. S. S., Kumar, N. A., Prasad, V. R., Venkataramana, V., Appalanaidu, S., Sridevi, B., Kumar, B. S. K., Bharati,
- M. D., Subbaiah, C. V., Acharyya, T., Rao, G. D., Viswanadham, R., Gawade, L., Manjary, D. T., Kumar, P. P., Rajeev, K., Reddy, N. P. C., Sarma, V. V., Kumar, M. D., Sadhuram, Y., and Murty, T. V. R.: High CO₂ emissions from the tropical Godavari estuary (India) associated with monsoon river discharges, Geophys. Res. Lett., 38, L08601, doi:10.1029/2011GL046928, 2011.

Sarma, V. V. S. S., Viswanadham, R., Rao, G. D., Prasad, V. R., Kumar, B. S. K., Naidu, S. A., Kumar, N. A., Rao, D. B.,

 Sridevi, T., Krishna, M. S. R., Reddy, N. P. C., Sadhuram, Y., and Murty, T. V. R.: Carbon dioxide emissions from Indian monsoonal estuaries, Geophys. Res. Lett., 39, L03602, doi: 10.1029/2011GL050709, 2012.

- Sarma, V. V. S. S., Paul, S., Vani, G. D., and Murty, V. S. N.: Impact of river discharge on the coastal water pH and pCO₂ levels during the Indian Ocean Dipole (IOD) years in the western Bay of Bengal, Cont. Shelf Res., 107, 132–140, doi:10.1016/j.csr.2015.07.015, 2015.
- 880 Schlünz, B., and Schneider, R. R.: Transport of terrestrial organic carbon to the oceans by rivers: re-estimating flux- and burial rates, Int. J. Earth Sci., 88, 599–606, doi:10.1007/s005310050, 2000.
 - Schmidt, C., Krauth, T., and Wagner, S.: Export of plastic debris by rivers into the sea, Environ. Sci. Technol., 51, 12246–12253, doi:10.1021/acs.est.7b02368, 2017.
 - Shi, W., Chen, Q., Yi, Q., Yu, J., Ji, Y., Hu, L., and Chen, Y.: Carbon emission from cascade reservoirs: Spatial
- heterogeneity and mechanisms, Environ. Sci. Technol., 51, 12175–12181, doi:10.1021/acs.est.7b03590, 2017.
 Stanford, J. A., and Ward, J. V.: Revisiting the serial discontinuity concept, River Res. Appl., 17, 303–310, doi:10.1002/rrr.659, 2001.

- Stanley, E. H., Powers, S. M., Lottig, N. R., Buffam, I., and Crawford, J. T.: Contemporary changes in dissolved organic carbon (DOC) in human-dominated rivers: is there a role for DOC management?, Freshwater Biol., 57, 26–42, doi:10.1111/j.1365-2427.2011.02613.x, 2011.
- Syvitski, J. P. M., Vörösmarty, C. J., Kettner, A. J., and Green, P.: Impact of humans on the flux of terrestrial sediment to the global coastal ocean, Science, 308, 376–380, doi:10.1126/science.1109454, 2005.
 - Townsend, C. R.: Concepts in river ecology: pattern and process in the catchment hierarchy, Arch. Hydrobiol. (suppl.), 113, 3–21, doi:10.1127/lr/10/1996/3, 1996.
- 895 Vannote, R. L., Minshall, G. W., Cummins, K. W., Sedell, J. R., and Cushing, C. E.: The river continuum concept, Can. J. Fish. Aquat. Sci., 37, 130–137, doi:10.1139/f80-017, 1980.
 - Wang, S., Fu, B., Piao, S., Lü, Y., Ciais, P., Feng, X., and Wang, W.: Reduced sediment transport in the Yellow River due to anthropogenic changes, Nat. Geosci., 9, 38–41, doi:10.1038/ngeo2602, 2015.
 - Wang, X., He, Y., Yuan, X., Chen, H., Peng, C., Zhu, Q., Yue, J., Ren, H., Deng, W., and Liu, H.: pCO₂ and CO₂ fluxes of
- the metropolitan river network in relation to the urbanization of Chongqing, China, J. Geophys. Res. Biogeocsi., 122, 470–486, doi:10.1002/2016JG003494, 2017.

Ward, J. V., and Stanford, J. A.: Serial discontinuity concept of lotic ecosystems, In: Dynamics of Lotic Systems, Fontaine, T.D., and Bartell, S.M. (Eds.), Ann Arbor Science, Ann Arbor, 29–42, 1983.

Ward, N. D., Bianchi, T. S., Medeiros, P. M., Seidel, M., Richey, J. E., Keil, R. G., and Sawakuchi, H. O.: Where Carbon

- 905 Goes When Water Flows: Carbon Cycling across the Aquatic Continuum, Front. Mar. Sci., 4, 7, doi:10.3389/fmars.2017.00007, 2017.
 - Webster, J. R.: Spiraling down the river continuum: stream ecology and the U-shaped curve, J. N. Am. Benthol. Soc., 26, 375–389, doi:10.1899/06-095.1, 2007.

Wehrli, B.: Conduits of the carbon cycle, Nature, 503, 346–347, doi:10.1038/503346a, 2013.

- 910 Weyhenmeyer, G. A., Fröberg, M., Karltun, E., Khalili, M., Kothawala, D., Temnerud, J., and Tranvik, L. J.: Selective decay of terrestrial organic carbon during transport from land to sea, Glob. Change Biol., 18, 349–355, doi:10.1111/j.1365-2486.2011.02544.x, 2012.
 - Winemiller, K. O., McIntyre, P. B., Castello, L., Fluet-Chouinard, E., Giarrizzo, T., Nam, S., Baird, I. G., Darwall, W., Lujan, N. K., Harrison, I., Stiassny, M. L. J., Silvano, R. A. M., Fitzgerald, D. B., Pelicice, F. M., Agostinho, A. A.,
- Gomes, L. C., Albert, J. S., Baran, E., Petrere Jr., M., Zarfl, C., Mulligan, M., Sullivan, J. P., Arantes, C. C., Sousa, L. M., Koning, A. A., Hoeinghaus, D. J., Sabaj, M., Lundberg, J. G., Armbruster, J., Thieme, M. L., Petry, P., Zuanon, J., Torrente Vilara, G., Snoeks, J., Ou, C., Rainboth, W., Pavanelli, C. S., Akama, A., van Soesbergen, A., and Sáenz L.: Balancing hydropower and biodiversity in the Amazon, Congo, and Mekong, Science, 351, 128–129, doi:10.1126/science.aac7082, 2016.
- 920 Wit, F., Muller, D., Baum, A., Warneke, T., Pranowo, W. S., Muller, M., and Rixen, T.: The impact of disturbed peatlands on river outgassing in Southeast Asia, Nat. Commun., 6, 10155, doi:10.1038/ncomms10155, 2015.
 - Yao, G., Gao, Q., Wang, Z., Huang, X., He, T., Zhang, Y., Jiao, S., and Ding, J.: Dynamics of CO₂ partial pressure and CO₂ outgassing in the lower reaches of the Xijiang River, a subtropical monsoon river in China, Sci. Total Environ., 376, 255–266, doi:10.1016/j.scitotenv.2007.01.080, 2007.
- 925 Yoon, T. K., Jin, H., Oh, N.-H., and Park, J.-H.: Technical note: Assessing gas equilibration systems for continuous pCO₂ measurements in inland waters, Biogeosciences, 13, 3915-3930, doi:10.5194/bg-13-3915-2016, 2016.

Yoon, T. K., Jin, H., Begum, M. S., Kang, N., and Park, J.-H.: CO₂ outgassing from an urbanized river system fueled by wastewater treatment plant effluents, Environ. Sci. Technol., 51, 10459–10467, doi:10.1021/acs.est.7b02344, 2017.

Zhai, W., Dai, M., Cai, W. J., Wang, Y., and Wang, Z.: High partial pressure of CO₂ and its maintaining mechanism in a

- 930 subtropical estuary: The Pearl River estuary, China, Mar. Chem., 93, 21–32, doi:10.1016/j.marchem.2004.07.003, 2005.
 Zhang, S. R., Lu, X. X., Higgitt, D. L., Chen, C. T. A., Sun, H. G., and Han, J. T.: Water chemistry of the Zhujiang (Pearl
 - River): Natural processes and anthropogenic influences, J. Geophys. Res. Earth Surf., 112, F01011, doi:10.1029/2006JF000493, 2007.
 - Zhang, S., Lu, X. X., Sun, H., Han, J., and Higgitt, D. L.: Major ion chemistry and dissolved inorganic carbon cycling in a
- human-disturbed mountainous river (the Luodingjiang River) of the Zhujiang (Pearl River), China, Sci. Tot. Environ.,
 407, 2796–2807, doi: 10.1016/j.scitotenv.2008.12.036, 2009.
 - Zhang, L. J., Wang, L., Cai, W.-J., Liu, D. M., and Yu, Z. G.: Impact of human activities on organic carbon transport in the Yellow River, Biogeosciences, 10, 2513–2524, doi:10.5194/bg-10-2513-2013, 2013.
 - Zhang, H., Liu, S., Yuan, W., Dong, W., Xia, J., Cao, Y., and Jia, Y.: Loess Plateau check dams can potentially sequester eroded soil organic carbon, J. Geophys. Res. Biogeosci., 121, 1449–1455, doi:10.1002/2016JG003348., 2016.

940

Zou, J.: Sources and dynamics of inorganic carbon within the upper reaches of the Xi River basin, Southwest China. PLoS ONE 11(8), e0160964, doi:10.1371/journal. pone. 0160964, 2016.

River	Receiving sea	Region	Basin area (10 ³ km ²)	Annual discharge (km ³ yr ⁻¹)	Population (×10 ⁶)	Population density (per km ²)	Annual discharge per capita (m ³ yr ⁻¹)
Ganges	Bay of Bengal	S Asia	980	490	411	419	1193
Brahmaputra	Bay of Bengal	S Asia	670	630	145	216	4353
Indus	Arabian Sea	S Asia	980	5 (90)	220	224	23 (410)
Krishna	Bay of Bengal	S Asia	260	12 (62)	101	390	118 (611)
Godavari	Bay of Bengal	S Asia	310	92 (120)	121	390	761 (993)
Mekong	South China Sea	SE Asia	800	550	64	80	8594
Yellow	Yellow Sea	E Asia	750	15 (43)	120	160	125 (358)
Yangtze	East China Sea	E Asia	1800	900	475	264	1894
Pearl River	South China Sea	E Asia	490	260	95	193	2749
Han River	Yellow Sea	E Asia	25	17	13	513	1326
Asia total			32518	11000	4835	148	227
			(32518)	(13196)			
Global total			105000	36000	7345	70	4901
			(106326)	(38170)			

Table 1. Geographic and demographic features of the major river systems addressed in the review, in comparison with Asian and global sums.

- 945 River basin area and discharge data were obtained from Milliman and Farnsworth (2011), supplemented with Asian and global sums in parentheses from Ludwig et al., (1996). Pre-diversion discharge data are provided in parentheses for the rivers where discharge has substantially decreased in recent years because of river diversion, reservoir construction, and irrigation. The Asia total discharge provided by Milliman and Farnsworth (2011) was estimated for all rivers of Asia and Oceania, excluding Arctic rivers in Russia. Population data were taken from various sources including Schmidt et al. (2017) and CIA World Factbook (https://www.cia.gov/library/publications/the-world-factbook/index.html/; last
- 950 accessed on 10 January 2018). Asia total population is the population for all countries belonging to Asia and Oceania excluding Russia.

Table 2. Summary of water use and wastewater production in southern and eastern Asia. Source of data: AQUASTAT, aglobalwaterinformationsystemoperatedbyFoodandAgricultureOrganization(FAO;http://www.fao.org/nr/water/aquastat/sets/index.stm).

Country	Population	Renewable surface water	Dam capacity		Municipal water		Municipal wastewater			
	(2015)	(2014)					Produced		Treated	
	×10 ³	km ³ yr ⁻¹	km ³	Year	km ³ yr ⁻¹	Year	km ³ yr ⁻¹	Year	km ³ yr ⁻¹	Year
Bangladesh	160996	1206.0	6.5	2013	3.6	2008	0.7	2000		
Bhutan	775	78.0			0.0	2008	0.0	2000		
Brunei	423	8.5	0.0	2010	0.2	2009				
Cambodia	15578	471.5			0.1	2006			0.0	1994
China	1407306	2739.0	829.8	2013	75.0	2013	48.5	2013	49.3	2014
North Korea	25155	76.2	13.6	2015	0.9	2005				
India	1311051	1869.0	224.0	2005	56.0	2010	15.5	2011	4.4	2011
Indonesia	257564	1973.0	23.0	2015	14.0	2005	14.3	2012		
Japan	126573	420.0	29.0	1993	15.4	2009	16.9	2011	11.6	2011
Laos	6802	333.5	7.8	2010	0.1	2003	0.1	2008	0.0	1995
Malaysia	30331	566.0	22.5	2015	3.9	2005	4.2	2009	2.6	2009
Mongolia	2959	32.7	0.3	2015	0.1	2009	0.1	2012	0.1	2006
Myanmar	53897	1157.0	15.5	2005	3.3	2000			0.0	1995
Nepal	28514	210.2	0.1	2015					0.0	2006
Pakistan	188925	239.2	27.8	2015	9.7	2008	3.1	2011	0.0	2002
Papua New Guinea	7619	801.0	0.7	2010	0.2	2005				
Philippines	100699	444.0	6.3	2006	6.2	2009	1.3	2011		
South Korea	50293	67.1	16.2	1994	6.9	2005	7.8	2011	6.6	2011
Singapore	5619		0.1	2015	1.1	2005	0.5	2013	0.5	2013
Sri Lanka	20715	52.0	5.9	1996	0.8	2005	0.1	2009		
Thailand	67959	427.4	68.3	2010	2.7	2007	5.1	2012	1.2	2012
Timor	1185	8.1			0.1	2004				
Vietnam	93448	847.7	28.0	2010	1.2	2005	2.0	2012	0.2	2012
S/SE/E Asia	3964386	14027.1	1325.2		201.6		120.2		76.5	
World	7344837	52952.7	7039.6		464.1		311.6		187.1	

River system		Mean	Method	Reference			
	Basin-wide	Mainstem	Headwater	Tributary	Impoundment		
Global	3100					Е	Raymond et al., 2013;
	(0-100000)						GLORCIH ^a
	2400					Е	Lauerwald et al., 2015;
	(2019–2826)						GLORICH ^a
			1116 ^b			Е	Marx et al., 2017;
			(0-97906)				GLORICH ^a
Asia (total)	1747	1255	973	2118	835		Sum of freshwater data below
	(28-11793)	(35-10977)	(74-5076)	(28-11793)	(128-8785)		
S. Asia							
Ganges	893	1083	401	1685	181, 224°	E/M	Manaka et al., 2015; GLORICH ^a
	(65-2620)	(65-2184)	(165-1222)	(1035-2620)			
Brahmaputra	664	494	292	758		Е	Huang et al., 2011; Manaka et al.
	(28-6706)	(65-6706)	(208-513)	(28-3678)			2015; Qu et al., 2017; GLORICH ^a
Indus	853	941		768		Е	GLORICH ^a
	(117-7725)	(117-7725)		(165-3161)			
Krishna	2152	1871		2305		Е	GLORICH ^a
	(711-4098)	(976-2536)		(711-4098)			
Godavari	8785 ^d				8785 ^d	Е	Prasad et al., 2013
Bhote Kosi	592	592				Е	GLORICH ^a
	(35-5907)	(35-5907)					
Various	2685	3081			609	E/M	Panneer Selvam et al., 2014
	(420-10977)	(426-10977)			(420-692)		
Cochin	(2975-6001) ^e					Е	Gupta et al., 2009 (saline estuary)
Other	5882					Е	Sarma et al., 2012 (saline estuary)
estuaries	(293-18492)						

Table 3. Summary of pCO_2 measured (M) or estimated (E) for the rivers in South (S.), Southeast (S.E.), and East (E.) Asia in comparison with data for the global rivers including Asian rivers. Only a single value per site was used to obtain the mean and range of each river system, so multiple measurements at a site were averaged to provide one representative value.

S.E. Asia							
Mekong	1235	1120	1367 ^f	1310	882 ^g	E/M	Alin et al., 2011; Li et al., 2013;
	(110-4503)	(703-1687)		(110-4503)	(864-899)		Manaka et al., 2015b
Red River	1589	1589				Μ	Le et al., 2017
	(992-3129)	(992-3129)					
Irrawaddy	2007	2007				E	Manaka et al., 2015b
	(1222-3157)	(1222-3157)					
Chao Phraya	3202	2814	3357			E	Manaka et al., 2015b
	(925-5076)	(2632-2996)	(925-5076)				
Malaysian	1217	1217				Μ	Wit et al., 2015
Rivers	(1159-1274)	(1159-1274)					
Indonesian	5262	5262				Μ	Wit et al., 2015
Rivers	(2400-8555)	(2400-8555)					
E. Asia							
Yellow	2164	2104	1083	2470	555, 441 ^h	E/M	Ran et al., 2015a; Ran et al., 2017a
	(147-9659)	(582-4770)	(147-3546)	(425-9659)	(266-735)		
Yangtze	2355	1202	565	2673	1393	Е	Qu et al., 2015; Liu et al., 2016; Ran
	(74-7718)	(528-2405)	(74-3465)	(249-7718)	(760-1908)		et al., 2017b ⁱ ; GLORICH ^a
Pearl River	1805	2465	1701	1967		Е	Yao et al., 2007; Zhang et al., 2009;
	(231-5414)	(2136-2833)	(231-5414)	(282-4808)			Zou, 2016; GLORICH ^a
Han River	2571	1480	628	4670	251	М	Yoon et al., 2017
	(101-11793)	(173-4089)		(101-11793)	(128-454)		

^aGLORICH: Global River Chemistry Database (Hartmann et al., 2014), from which Raymond et al. (2013) and Lauerwald et al. (2015) calculated pCO_2 for global rivers, but excluded data obtained at pH <5.4 and at pH <5.4 and high pollution levels, respectively. Marx et al. (2017) excluded pCO_2 values above 100,000 µatm to avoid any potential overestimation resulting from pH and alkalinity effects; ^bThe mean was calculated from the global mean pCO_2 (3100 µatm; Raymond et al., 2013) by assuming that headwaters account for 36% of the global riverine CO₂ emission (Marx et al., 2017); ^cDakpatthar Barrage on the Yamuna, a Ganges tributary (cf. the other barrage at Rishkesh on the mainstem)

5 using headspace equilibration method (Park, unpublished data); ^dDowleiswaram Reservoir (only the mean of time-series data was included in calculating the regional mean and range); ^eCochin estuary: two sites on the Periyar River and 11 sites on the estuary; ^fMeasured at Lancang headwater at Qinghai, China using headspace equilibration method (Park, unpublished data); ^gImpounded tributaries of the Lower Mekong (Li et al., 2013); ^hImpoundments on a Yellow River tributary (Ran et al., 2017a),; ⁱRan et al. (2017b) excluded *p*CO₂ values calculated at pH <6.5.</p>

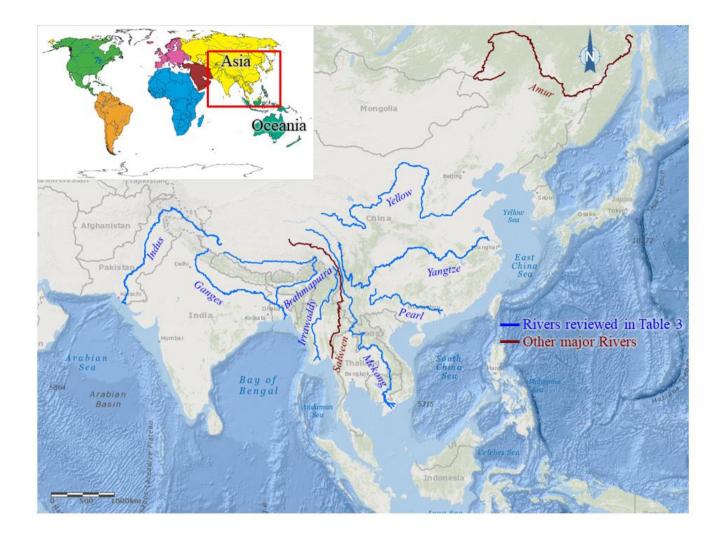


Figure 1. Major river systems of South, Southeast, and East Asia that belong to top 30 global rivers based on discharge (Raymond and Spencer, 2015). The base map and the inset world map were modified from ArcGIS online Ocean Basemap
and Milliman and Farnsworth (2011), respectively. Rivers addressed in the review and other large rivers are distinguished by different colors. Three Asian regions comprise the majority of Asian countries included in the regional categories "Asia" (indicated by a yellow color on the inset world map) and "Oceania" (dark green) used by Milliman and Farnsworth (2011).

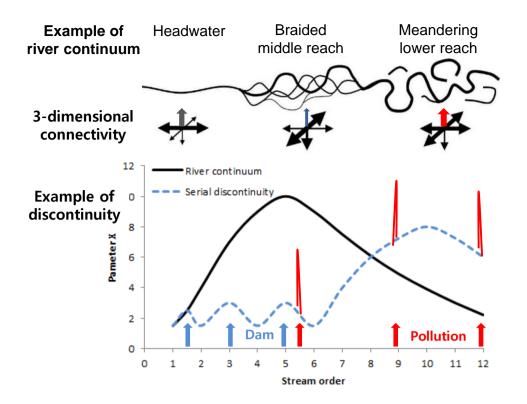


Figure 2. A schematic diagram describing river discontinuity in the anthropogenic land-water-scape: an example of river continuum observed in a minimally impacted river (top); three-dimensional connectivity vectors along the up-, mid-, and
lower river reaches (middle); longitudinal variations in a hypothetical riverine biogeochemical process X (bottom). The connectivity vectors and the plots depicting river continuum and serial discontinuity were modified from Stanford and Ward (2001) and Poole (2010), complemented with some additional considerations including pollution-induced pulsatile discontinuities (red-colored pulses) and the vertical vector of air-water gas exchange (blue and red color indicating potential effects of dams and water pollution, respectively).

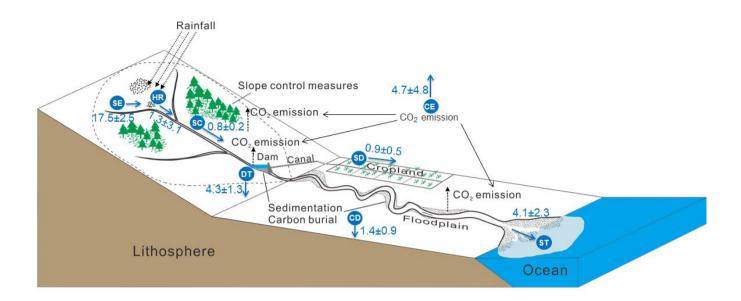


Figure 3. A schematic diagram illustrating human-induced alterations of the riverine C fluxes in the Yellow River as a model river system (modified from Ran et al., 2014). The annual flux rate (Tg C yr⁻¹) for each of the described soil and

5 fluvial processes was estimated for the period 1950 – 2010. Slope and fluvial processes depicted in the figure include – SE: soil erosion, HR: hillslope redistribution, SC: slope control of erosion, DT: dam trapping, SD: sediment diversion, CD: channel deposition, CE: CO₂ emission, and ST: seaward transport. Refer to Ran et al. (2014) for more details on their flux and uncertainty estimations.

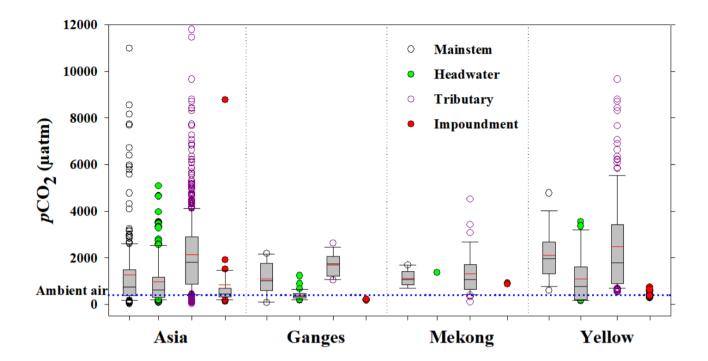


Figure 4. Comparison of pCO₂ measured or estimated for major Asian rivers including three rivers selected for a detailed review – the Ganges, Mekong, and Yellow River. The blue dotted line indicates the ambient air pCO₂ level around 400 atm. The horizontal black and red lines of the box plots are the median and mean value, respectively. Each box covers the 25th to 75th percentile, whereas the whiskers represent the 10th and 90th percentile. The number of data points included in the total Asian rivers: 1209 (mainstem: 261, headwater: 161, tributary: 757, impoundment: 30); the Ganges: 63 (mainstem: 14, headwater: 30, tributary: 17, impoundment: 2); the Mekong: 59 (mainstem: 19, headwater: 1, tributary: 37, impounded

tributary: 2), the Yellow River: 215 (mainstem: 14, headwater: 22, tributary: 164, impoundment: 15). Refer to Table 3 for

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more details on data sources.

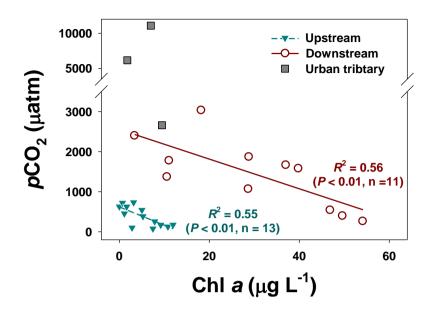


Figure 5. Longitudinal shift in the relationship between Chl a and pCO_2 observed between the up- and downstream reaches of the Han River receiving highly polluted urban tributaries (modified from the supplementary information of Yoon et al., 2017).