Supplementary Material

**Contents of this file**

Table S1 to S2

Figures S1 to S4

Table S1. Sampling sites and geographic information of the Ganges, Mekong, Yangtze, Yellow, and Han River.

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| **River** | **Site ID** | **Site name** | **Coordinates** | |
| **Mainstem** |  |  | **Latitude** | **Longitude** |
| Yamuna | Y1 | Yamunotri | 30°58'40.14"N | 78°26'26.10"E |
| Yamuna | Y2 | Dakpatthar | 30°30'3.66"N | 77°47'51.18"E |
| Yamuna | Y3 | Delhi | 28°36'2.16"N | 77°15'39.30"E |
| Yamuna | Y4 | Agra | 27°10'34.32"N | 78° 2'38.22"E |
| Ganges | G1 | Gangotri | 30°59'38.70"N | 78°56'39.24"E |
| Ganges | G2 | Alaknanda mouth | 30° 8'43.98"N | 78°35'52.56"E |
| Ganges | G3 | Bhagirathi mouth | 30° 8'22.08"N | 78°35'51.24"E |
| Ganges | G4 | Devprayag | 30° 7'43.56"N | 78°35'48.54"E |
| Ganges | G5 | Rishikesh | 30° 7'43.56"N | 78°19'48.18"E |
| Ganges | G6 | Rishikesh | 30° 4'34.44"N | 78°17'14.52"E |
| Ganges | G7 | Kanpur | 26°25'3.54"N | 80°25'41.10"E |
| Ganges | G8 | Allahabad | 25°25'31.02"N | 81°53'17.34"E |
| Ganges | G9 | Varanasi | 25°18'8.22"N | 83° 0'34.56"E |
| Ganges | G10 | Patna | 25°36'51.24"N | 85°12'8.10"E |
| Ganges | G11 | Farakka | 24°48'51.24"N | 87°55'2.58"E |
| Hooghly | H1 | Kolkata | 22°33'9.48"N | 88°18'3.18"E |
| Hooghly | H2 | Diamond Harbor | 22°11'4.44"N | 88°11'21.78"E |
| Hooghly | H3 | Kakdwip | 21°52'49.80"N | 88° 9'50.40"E |
| Padma | G12 | Kustia | 24° 2'29.89"N | 89° 1'41.52"E |
| Padma | G13 | Maowa | 23°28'20.40"N | 90°15'19.95"E |
| Padma | G14 | Chandpur | 23°14'4.05"N | 90°38'42.32"E |
| Mekong | M1 | Jinghong | 22° 2'0.54"N | 100°47'0.30"E |
| Mekong | M2 | Jinghong | 21°59'52.20"N | 100°50'40.32"E |
| Mekong | M3 | Chaiang | 20°15'15.54"N | 100°10'41.34"E |
| Mekong | M4 | Phnom Penh | 11°43'38.03"N | 104°58'23.09"E |
| Mekong | M5 | Phnom Penh | 11°35'6.43"N | 104°56'37.21"E |
| Mekong | M6 | Phnom Penh | 11°32'50.10"N | 104°58'11.10"E |
| Mekong | M7 | Phnom Penh | 11°31'56.17"N | 104°55'56.03"E |
| Mekong | M8 | Can Tho | 10° 9'4.56"N | 105°39'13.80"E |
| Mekong | M9 | Can Tho | 10° 7'45.00"N | 105°41'6.36"E |
| Mekong | M10 | Can Tho | 10° 1'11.10"N | 105°48'30.18"E |
| Yellow | Y1 | Hancheng | 35°39'35.16"N | 110°35'47.64"E |
| Yellow | Y2 | Hancheng | 34°49'55.08"N | 111°20'17.64"E |
| Yellow | Y3 | Hohhot | 34°54'19.32"N | 113°40'40.08"E |
| Yellow | Y4 | Hohhot | 36°43'26.10"N | 116°59'2.28"E |
| Yellow | Y5 | Hohhot | 40°15'48.66"N | 111° 4'26.28"E |
| Yangtze | YR1 | Chongqing | 29°35'10.8"N | 106°34'30.84"E |
| Yangtze | YR2 | Three Goreges Dam | 30°49'36.04"N | 111° 3'5.75"E |
| Han River | H1 | Haean | 38°14'60.00"N | 128° 6'60.00"E |
| Han River | H2 | Mandae | 38°15'60.00"N | 128° 8'60.00"E |
| Han River | H3 | Soyang River | 38° 0'0.01"N | 128° 6'0.00"E |
| Han River | H4 | Jamsil | 37° 31.116' N | 127° 04.887' E |
| Han River | H5 | Noduel | 37° 31.083' N | 126° 57.598' E |
| Han River | H6 | Soyang Dam | 37°56'0.00"N | 127°49'0.00"E |
| Han River | H7 | Eoam Dam | 37°51'60.00"N | 127°40'60.00"E |
| Han River | H8 | Cheongpyeong | 37°43'0.00"N | 127°24'0.00"E |
| Han River | H9 | North Han | 37°36'0.00"N | 127°20'0.00"E |
| Han River | H10 | South Han | 37°31'0.00"N | 127°21'60.00"E |
| Han River | H11 | Paldang Dam | 37°30'0.00"N | 127°18'0.01"E |
| Han River | H12 | Haengju bridge | 37° 36.121' N | 126° 48.763' E |
| Han River | H13 | Amsa | 37°32'59.98"N | 127° 7'0.08"E |
| Han River | H14 | Bamseom | 37°32'0.00"N | 126°55'0.00"E |
| Han River | H15 | Bukhan | 37°32'17.00"N | 127°18'28.00"E |
| Han River | H16 | Jeonryuri | 37°40'59.99"N | 126°39'0.01"E |
| **Tributary** |  |  |  |  |
| Ganges | T1 | Brahmaputra, Sirajgangj | 24°23'6.13"N | 89°48'8.27"E |
| Ganges | T2 | Meghna, Sonargaon | 23°38'58.39"N | 90°37'31.88"E |
| Ganges | T3 | Buriganga, Dhaka | 23°41'55.98"N | 90°25'2.94"E |
| Ganges | T4 | Buriganga, Munshiganj | 23°34'16.08"N | 90°30'30.96"E |
| Ganges | T5 | Upstream of Buriganga-Meghna confluence, Munshiganj | 23°35'37.68"N | 90°28'28.51"E |
| Ganges | T6 | Meghna, Shikarikandi | 23°32'56.03"N | 90°34'45.50"E |
| Ganges | T7 | Meghna, Nayanagar | 23°20'47.46"N | 90°36'43.26"E |
| Mekong | T8 | Chaiang | 20°13'38.22"N | 100° 7'45.24"E |
| Mekong | T9 | Tonle Sap, Phnom Penh | 11°40'5.70"N | 104°51'40.14"E |
| Mekong | T10 | Tonle Sap, Phnom Penh | 11°35'21.48"N | 104°55'17.22"E |
| Mekong | T11 | Phnom Penh | 11°34'3.65"N | 104°56'0.13"E |
| Mekong | T12 | Can Tho | 9°56'53.22"N | 105°43'20.04"E |
| Mekong | T13 | Can Tho | 10° 0'21.96"N | 105°45'44.70"E |
| Mekong | T14 | Can Tho | 10° 1'39.60"N | 105°47'16.56"E |
| Yellow | T15 | DaHeiHe tributary | 40°15'29.40"N | 111° 8'53.82"E |
| Yellow | T16 | Wuding River, Hengshan-12 | 37°45'19.82"N | 109°12'16.14"E |
| Yellow | T17 | Wuding River, Hengshan-8 | 37°53'32.33"N | 109°16'17.44"E |
| Yellow | T18 | Wuding River, Zizhou-7 | 37°44'14.91"N | 109°54'9.57"E |
| Yellow | T19 | Wuding River, Zizhou-9 | 37°44'52.29"N | 109°53'47.45"E |
| Yellow | T20 | Wuding River, Wuding-2 | 38° 2'2.49"N | 109°37'2.31"E |
| Yellow | T21 | Wuding River, Hengshan-10 | 37°46'8.84"N | 109°11'11.76"E |
| Yellow | T23 | Wuding River, Hengshan-16 | 37°43'53.80"N | 109°12'10.48"E |
| Yellow | T24 | Wuding River, Zizhou-5 | 37°43'24.72"N | 109°54'47.02"E |
| Yellow | T25 | Wuding River, Weijialou-1 | 37°33'21.11"N | 109°32'9.12"E |
| Yellow | T26 | Wuding River, Yulin-17 | 38° 7'41.28"N | 109°46'14.33"E |
| Yellow | T27 | Wuding River, Yulin-18 | 38° 0'2.84"N | 109°49'28.85"E |
| Yellow | T28 | Wuding River, Hengshan-2 | 38° 1'13.58"N | 109° 4'6.77"E |
| Yellow | T29 | Wuding River, Hengshan-17 | 37°44'23.05"N | 109° 9'10.32"E |
| Yellow | T30 | Wuding River, Suide-1 | 37°33'44.98"N | 110°10'19.24"E |
| Yellow | T31 | Wuding River, Suide-2 | 37°26'49.11"N | 110°18'27.08"E |
| Yellow | T32 | Wuding River, Suide-3 | 37°22'29.81"N | 110°21'51.73"E |
| Yellow | T33 | Wuding River, Suide-4 | 37°17'39.40"N | 110°24'21.51"E |
| Yellow | T34 | Wuding River, Suide-5 | 37°16'19.24"N | 110°27'39.74"E |
| Han River | T35 | Downstream of Hongjecheon | 37° 33.605' N | 126° 53.744' E |
| Han River | T36 | Downstream of Jungnangcheon | 37° 33.185' N | 127° 02.800' E |
| Han River | T37 | Upstream of Jungnangcheon | 37° 48.338' N | 127° 01.897' E |
| Han River | T38 | Midstream of Jngnangcheon | 37°41.462' N | 127°02.987' E |
| Han River | T39 | Upstream of Godeokcheon | 37° 31.880' N | 127° 11.100' E |
| Han River | T40 | Downstream of Godeokcheon | 37° 34.039' N | 127° 09.738' E |
| Han River | T41 | Upstream of Tancheon | 37°26.912' N | 127°04.697' E |
| Han River | T42 | Upstream of Tancheon | 37° 22.243' N | 127° 02.088' E |
| Han River | T43 | Downstream of Tancheon | 37° 30.499' N | 127° 04.134' E |
| Han River | T44 | Upstream of Anyangcheon | 37° 27.033' N | 126° 56.927' E |
| Han River | T45 | Downstream of Anyangcheon | 37° 32.861' N | 126° 52.897' E |
| Han River | T46 | Upstream of Hongjecheon | 37° 35.911' N | 126° 58.216' E |
| Han River | T47 | Upstream of Changneungcheon | 37° 40.753' N | 126° 57.681' E |
| Han River | T48 | Downstream of Changneungcheon | 37° 35.781' N | 126° 49.985' E |
| **Wastewater** |  |  |  |  |
| Ganges | W1 | Buriganga, Dhaka | 23°49'24.24"N | 90°15'29.34"E |
| Ganges | W2 | Buriganga, Dhaka | 23°47'17.45"N | 90°14'31.01"E |
| Ganges | W3 | Buriganga, Dhaka | 23°50'45.06"N | 90°14'33.66"E |
| Ganges | W4 | Tolly Canal mouth, Kalkata | 22°32'52.92"N | 88°19'29.70"E |
| Mekong | W5 | Can Tho | 10° 2'16.56"N | 105°47'23.40"E |
| Mekong | W6 | Can Tho | 9°57'30.42"N | 105°43'32.28"E |
| Mekong | W7 | Can Tho | 10° 0'12.42"N | 105°44'40.44"E |
| Mekong | W8 | Tonle Sap, Phnom Penh | 11°39'35.57"N | 104°51'39.35"E |
| Mekong | W9 | Tonle Sap, Phnom Penh | 11°38'23.17"N | 104°53'2.83"E |
| Mekong | W10 | Tonle Sap, Phnom Penh | 11°34'39.11"N | 104°55'32.52"E |
| Han River | W11 | Jungnang wastewater treatment plant, Seoul | 37° 33.176' N | 127° 03.888' E |

Table S2. Summary of bicarbonate ions (HCO3–) or dissolved inorganic carbon (DIC), NH4+, NO3–, and PO43– concentrations measured in five river systems in Asia between monsoon and dry season. Values are means followed by ranges in parentheses.

|  |  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
| Rivers | HCO3–/DIC (μmol L–1) | | NH4+ (mg L–1) | | NO3–(mg L–1) | | PO43–(mg L–1) | | References |
|  | Dry | Monsoon | Dry | Monsoon | Dry | Monsoon | Dry | Monsoon |  |
| Ganges | 3043  (1004–4536) | 1650  (894–2531) | 6.1  (0.04–27.7) | 2.6  (0.1–13.0) | 1.6  (0.1–5.9) | 1.6  (0.3–4.6) | 3.3  (1.1–5.9) | 0.5  (0.2–1.1) | Begum et al., 2021; Sarin et al., 1989 |
| Mainstem |  |  | 4.8  (0.1–27.7) | 4.0  (0.1–13.0) | 0.8  (0.1–1.7) | 1.9  (0.7–4.6) | 4.8 | 0.6  (0.2–1.1) |  |
| Tributary |  |  | 4.8  (0.04–14.3) | 0.3  (0.2–0.4) | 2.5  (0.2–5.9) | 1.1  (0.6–1.8) | 2.5  (1.1–3.8) | 0.3 |  |
| Wastewater |  |  | 8.9  (6.0–14.0) | 0.2  (0.1–0.4) | 2.5  (0.3–5.9) | 0.6  (0.3–0.8) | 3.3  (1.4–5.9) | 0.3 |  |
| Mekong |  | 1763  (1584–1943) | 9.7  (0.1–45.7) | 0.3  (0.1–0.9) | 1.7  (0.4–3.0) | 1.6  (1.0–2.2) | 1.7  (0.3–3.9) | 0.8  (0.04–1.6) | Begum et al., 2021; Manaka et al., 2015 |
| Mainstem |  |  | 0.1  (0.1–0.2) | 0.1  (0.1–0.2) | 1.3  (0.6–2.3) | 1.6  (1.0–2.7) |  |  |  |
| Tributary |  |  | 0.1  (0.09–0.1) | 0.1  (0.1–0.2) | 1.4  (0.4–2.8) | 1.4  (1.2–1.7) | 0.3 | 0.3 |  |
| Wastewater |  |  | 17.4  (0.8–45.7) | 0.5  (0.1–1.0) | 2.1  (0.4–3.0) | 1.6  (1.0–2.2) | 2.0  (0.3–3.9) | 0.8  (0.04–1.6) |  |
| Yellowa | 2570–3640 | 2269–2752 | 0.6 | 0.1 | 14.5  (6.9–18.4) | 14.2  (8.4–21.9) |  |  | Begum et al., 2021; Zhang and Zhang, 2007; Ran et al., 2013 |
| Mainstem |  |  | 0.6 | 0.1 | 14.5  (6.9–18.4) | 14.2  (8.4–21.9) |  |  |  |
| Tributary |  |  |  |  |  |  |  |  |  |
| Yangtzeb  Mainstem | 1330  (445–1816) |  |  |  |  | 7.7  (5.9–8.1) |  |  | Begum et al., 2021;  Chen et al., 2002 |
| Han River |  |  | 5.2  (0.01–51.1) | 1.2  (0.01–8.7) | 23.6  (8.2–49.4) | 18.2  (4.1–39.0) | 0.3 | 0.4  (0.2–1.0) | Jin et al., 2018; Unpublished data |
| Mainstem (U) |  |  | 3.3  (0.02–15.1) | 0.1  (0.07–0.14) | 10.4  (8.2–12.5) | 8.1  (4.1–10.9) |  |  |  |
| Mainstem (M) |  |  | 4.7  (0.1–20.5) | 0.3  (0.1–0.5) | 16.0  (12.3–18.8) | 11.7  (8.6–13.9) |  |  |  |
| Mainstem (L) |  |  | 4.4  (0.3–27.8) | 0.3  (0.2–0.4) | 20.5  (16.8–25.8) | 19.8  (14.2–25.6) | 0.3 |  |  |
| Tributary |  |  | 6.2  (0.01–51.1) | 1.6  (0.01–5.2) | 34.6  (26.2–43.5) | 20.9  (12.1–32.9) |  | 0.3  (0.2–0.5) |  |
| Wastewater |  |  | 4.7  (2.4–10.9) | 5.4  (2.1–8.7) | 49.4 | 26.1  (13.3–39.0) |  |  |  |

Yangtze River included data only for the mainstem sites; U: upper reach; M: middle reach; L: lower reach.

‘a’ Yellow River represents the ranges of DIC concentrations between monsoon and dry season.

‘b’ Yangtze River represents the average of HCO3– ion concentrations from 1958–1990.

Chart, scatter chart

Description automatically generated

Figure S1. Relationship between the calculated *p*CO2 and pH (A, D), DOC (B, E), or TA (C, F) in the mainstems (M), tributaries (T), and polluted urban tributaries or wastewater effluents (W) of four large river systems during the monsoon and dry season. For the four large rivers 128 data are presented from Figure 2 (excluding 2 extreme values exceeding 25,000 µatm during the monsoon season), including the mainstem (M) (n: 29 for monsoon season; 24 for dry season), tributaries (T) (n: 29 for monsoon season; 30 for dry season) and wastewater (W) (n: 6 for monsoon season; 10 for dry season) from 2015 to 2019. The significant relationship (*p*<0.05) is indicated by a regression line through the plot. (Partial pressure of CO2: *p*CO2, dissolved organic carbon: DOC, total alkalinity: TA)

Chart

Description automatically generated

Figure S2. The percentages of *p*CO2 overestimation, calculated as 100 × (calculated *p*CO2 –measured*p*CO2)/ measured *p*CO2, as a function of pH (A, D), DOC (B, E), and TA (C, F) in the mainstems (M), tributaries (T), and polluted urban tributaries or wastewater effluents (W) of four large river (the Ganges, Mekong, Yangtze, and Yellow River) during the monsoon and dry season. For the four large rivers, 125 data are presented from Figure 2 (excluding 5 extreme values exceeding 500% during the monsoon season), including the mainstem (M) (n: 27 for monsoon season; 23 for dry season), tributaries (T) (n: 28 for monsoon season; 31 for dry season) and wastewater (W) (n: 6 for monsoon season; 10 for dry season) from 2015 to 2019. The significant relationship (*p*<0.05) is indicated by a regression line through the plot. (Partial pressure of CO2: *p*CO2, dissolved organic carbon: DOC, total alkalinity: TA)

Chart, scatter chart

Description automatically generated

Figure S3. Relationship between the calculated *p*CO2 and pH (A, D), DOC (B, E), or TA (C, F) in the mainstems (M), tributaries (T), and wastewater effluents (W) of the Han River during the monsoon and dry season. 212 data are presented from Figure 2 (excluding 6 extreme values exceeding 35,000 and 25,000 µatm during the monsoon and dry periods, respectively), including the mainstem (M) (n: 55 for monsoon season; 83 for dry season), tributaries (T) (n: 25 for monsoon season; 43 for dry season) and wastewater (W) (n: 2 for monsoon season; 4 for dry season) from 2014 to 2020. For the dry season, samples are indicated by filled symbols for the months without “rainfall”, while void symbols represent the months with “sporadic rainfall”. The significant relationship (*p*<0.05) is indicated by a regression line through the plot. (Partial pressure of CO2: *p*CO2, dissolved organic carbon: DOC, total alkalinity: TA)

Graphical user interface, chart, scatter chart

Description automatically generated

Figure S4. The percentages of *p*CO2 overestimation, calculated as 100 × (calculated *p*CO2 –measured*p*CO2)/ measured *p*CO2, as a function of pH (A, D), DOC (B, E), and TA (C, F) in the mainstems (M), tributaries (T), and wastewater effluents (W) of the Han River during the monsoon and dry season. 209 data are presented from Figure 2 (excluding 9 extreme values exceeding 2000% during the dry periods), including the mainstem (M) (n: 55 for monsoon season; 84 for dry season), tributaries (T) (n: 22 for monsoon season; 42 for dry season), and wastewater (W) (n: 2 for monsoon season; 4 for dry season) from 2014 to 2020. For the dry season, samples are indicated by filled symbols for the months without “rainfall”, while void symbols represent the months with “sporadic rainfall”. The significant relationship (*p*<0.05) is indicated by a regression line through the plot. (Partial pressure of CO2: *p*CO2, dissolved organic carbon: DOC, total alkalinity: TA)