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Conservative behavior of dissolved black carbon in the northwestern Pacific marginal seas

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Dissolved black carbon (DBC) represents the largest molecularly identifiable slow-cycling organic carbon pool in the ocean. However, its behavior remains debated due to large differences between its radiocarbon ages and residence times based on mass balance estimations, suggesting considerable removal in the source regions. Here, we show that DBC is predominantly derived from riverine sources and behaves conservatively throughout the entire water masses of the northwestern Pacific marginal seas—including the East China Sea, the Yellow Sea, and the East Sea (Japan Sea)—which are characterized by extreme biogeochemical alterations and long water residence times (~100 years). This conservative behavior is evidenced by a strong negative correlation between DBC and salinity, consistent mass balance estimates, and a uniform B6CA/B5CA marker ratio. Thus, we suggest that the discrepancy between DBC ages and residence times in the ocean is more likely due to substantially enhanced contemporary DBC production rather than removal pathways.

Black carbon (BC) is a mixture of condensed aromatic compounds, produced through incomplete combustion of biomass and fossil fuels[1]. Thermal alteration enhances its chemical stability, making it highly resistant to biodegradation and contributing to long-term carbon storage with important implications for the global carbon cycle[2,3]. Due to this recalcitrance, BC persists in the environment on timescales of centuries to millennia[4,5]. A fraction of BC can be mobilized and transported to the oceans in a dissolved form, referred to as dissolved BC (DBC), primarily through river runoff[6,7]. The annual flux of riverine DBC to the oceans is estimated to be 18 ± 4 Tg yr^{-1}, accounting for ~10% of the global riverine flux of dissolved organic carbon (DOC)[7,8]. In the global ocean, DBC is one of the most prevalent organic molecular classes quantified, constituting ~2% of the total oceanic DOC[9].

Assuming a steady-state in which global riverine DBC fluxes (18 Tg yr^{-1})[8] balance the oceanic DBC reservoir (14 Gt)[10], the estimated turnover time would be ~800 years. However, the average ^{14}C age of the oceanic DBC is estimated to be 4800 ± 620 ^{14}C years in the surface ocean and 23,000 ± 3000 ^{14}C years in the deep ocean[10]. Given that riverine DBC is of near-modern age[11], this discrepancy in the ages of DBC in the river and ocean implies that substantial removal needs to occur before DBC reaches the deep ocean if the input flux is constant over the last 20,000 years. Accordingly, although DBC is highly recalcitrant, previous studies in the East China Sea[12], south of Japan[12], western South China Sea[13], Bohai Sea[14], and Bering Sea[15] suggested removal processes via photodegradation in surface waters[16] and particle adsorption in surface and deep waters[9,17,18]. Furthermore, the stable carbon isotope values of oceanic DBC are ~6‰ enriched compared to those of riverine DBC, suggesting that rivers are not the predominant source of oceanic DBC[19]. To explain this, additional sources that have been suggested, yet poorly understood, include autochthonous production[19], hydrothermal vents[20], the contribution of petrogenic BC[21], and atmospheric deposition[22], assuming that the

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isotopic value remains unchanged. Nevertheless, some local studies in the Delaware Estuary[23], East China Sea[24], south Yellow Sea[24], and northern South China Sea[25] have reported dominant riverine inputs and conservative DBC behavior in the near-source regions. In the deep Southern Ocean, DBC was found to be conservative, indicating its salt-like stability[26]. Therefore, the behavior of DBC in the ocean should be further investigated in well-defined oceanic settings characterized by distinct sources, intense biogeochemical reactions, and long water residence times.

Thus, in this study, we aim to investigate the distribution, sources, and behavior of DBC in the northwestern Pacific marginal seas (the East China Sea, Yellow Sea, and East Sea), which are characterized by large shelf areas, deep waters, and long water residence times (~100 years). The continental shelf of the East China Sea (ECS) and the Yellow Sea is one of the largest continental shelves in the world, covering a total area of 9.5×10^5 km[2][27]. This shelf receives significant terrestrial material inputs through river discharge, notably from the Changjiang River (Yangtze River)[28], the third-largest river globally (annual water discharge = 0.9×10^{12} m^{-3} yr^{-1}), as well as through submarine groundwater discharge (SGD)[29]. Substantial amounts of terrestrial material are transported from the Asian continent through atmospheric deposition[30]. In addition, the biogeochemical properties (e.g., DOC and FDOM) of the Yellow Sea are known to be significantly altered during its residence time of ~5 years[29,31].

The waters of the Yellow Sea and the ECS, mixed with the Kuroshio Current, enter the East Sea (Japan Sea). The East Sea is a semi-enclosed marginal sea in the northwestern Pacific Ocean surrounded by Korea, Japan, and Russia, with a depth of up to 3500 m and an average depth of 1680 m. Its exchange with the North Pacific Ocean is limited through only shallow straits (<150 m), including the Tsugaru, Soya, and Korea Straits[32,33]. The East Sea has its own deep-water formation and meridional circulation[33,34] with a long turnover time scale of ~100 years[35]. The East Sea has been regarded as a miniature ocean with oceanic features such as large-scale current systems, subpolar fronts, mesoscale eddies, and deep water formations similar to the global ocean[36,37]. Thus, the study in this interconnected marginal sea system, compared with previous regional or global studies, provides a unique opportunity to examine DBC behavior under conditions of strong source inputs, intense biogeochemical and photochemical reactions in the semi-enclosed upper ocean, and removal processes in the fully enclosed deep sea (200–3000 m).

Results

Distributions of DBC

For the surface water of the entire study region, including the ECS, the Yellow Sea, and the East Sea, the concentrations of DOC and DBC ranged from 68 to 109 μM and from 1.18 to 3.54 μM, respectively (Fig. 1b, c). Both DOC and DBC concentrations in the surface water showed a decreasing trend from the ECS shelf to the East Sea. The ratio of benzenehexacarboxylic acid to pentacarboxylic acid (B6CA/B5CA), an indicator of molecular condensation of DBC[16], ranged from 0.25 to 0.30 (Fig. 1d). The ratios were relatively constant over the entire study region.

In the deep waters of the East Sea, the concentrations of DOC and DBC were 14% and 10% lower, respectively, than those in the surface waters and remained relatively constant with depth (Fig. 1e, f). The average concentrations of DOC and DBC were 70 ± 5 μM and 1.32 ± 0.11 μM in the surface layer (0–200 m), and 60 ± 3 μM and 1.19 ± 0.08 μM in the deep layer (>200 m), respectively. The B6CA/B5CA ratio was relatively uniform throughout the water column (average: 0.28 ± 0.02) (Fig. 1g).

Discussion

The DBC concentrations in the surface East Sea (0–200 m; 1.32 ± 0.11 μM) were comparable to those reported from other marginal seas, including the Canada Basin (1.4 ± 0.3 μM)[15], the Chukchi Sea (1.3 ± 0.2 μM)[15], and the South China Sea (1.0 ± 0.2 μM)[13], but they were higher than those observed in major oceans, including the Pacific (0.6 ± 0.1 μM)[18,19], the Atlantic (0.8 ± 0.0 μM)[19], and the Indian Oceans[9] (Supplementary Table 2). In the deep sea (>1000 m), DBC concentrations in the East Sea (1.19 ± 0.08 μM) were similar to those observed in the Sargasso Sea (1.2 ± 0.1 μM)[10], but they were notably higher than those reported from other marginal seas and open oceans[9,13,15,18,19,38]. Such elevated deep-water concentrations may be driven by the rapid turnover of the deep water in the East Sea, which has also been suggested as a possible cause of the high DOC concentrations observed in the region[39].

The plot of DOC concentrations versus salinity showed significant accumulation of DOC in the Yellow Sea relative to the two-endmember mixing line between the Changjiang diluted water and the Kuroshio Current water (Fig. 2a), as documented by Han et al.[40]. This accumulation was primarily attributed to benthic inputs, as supported by similarly elevated activities of ^{228}Ra (half-life: 5.75 years)[40], which is conservative in seawater. Accordingly, in the center of the Yellow Sea, which has a water residence time of ~5 years based on ^{228}Ra age[31], both ^{228}Ra activities (measured in the same samples as in a previous study[41]) and DOC concentrations were approximately twice as high as those observed in the ECS at comparable salinity, showing a significant correlation[40].

In contrast to the observed non-conservative behavior of DOC in the Yellow Sea, the plot of DBC concentrations versus salinity across the entire study region showed a significant negative correlation ($R^2 = 0.89$, $p < 0.001$, $n = 10$) (Fig. 2b), indicating that DBC is neither significantly added to nor removed from the system during transport from the ECS shelf to the East Sea. It is important to note that DBC was consistently conservative over the water residence time of ~5 years, based on ^{228}Ra activities. The intercept of the salinity–DBC plot (10.9 μM) falls within the range of DBC concentrations observed in the Changjiang River (2.3–14.3 μM)[42], indicating that riverine input is the primary source of the DBC in these marginal seas. This result is consistent with recent findings that rivers are the dominant source of DBC in coastal waters[14,15]. Since our dataset includes samples collected across different seasons and years, seasonal variability in hydrodynamics and river discharge could influence DBC distributions and composition, especially in low-salinity waters. However, such effects are likely averaged over the water residence times, and thus DBC concentrations should remain consistent in the aged waters of the Yellow Sea, as inferred from ^{228}Ra activities in the same samples.

Although riverine input appears to dominate, potential contributions from non-riverine sources cannot be ruled out. The atmospheric deposition accounts for 3–18% of the riverine input in this region (see the Methods). Bao et al.[24] reported that atmospheric deposition together with other unidentified sources contributed ~15% of oceanic DBC in the ECS during spring. Consistently, rainwater DBC concentrations in this region (0.02–4.79 μM)[43] are generally much lower than the freshwater endmember value inferred from our salinity–DBC plot (10.9 μM), indicating only a minor contribution of precipitation to DBC inputs. Previous studies have shown that SGD contributed ~12% of the riverine DBC flux in the Jiulong estuary[44], sedimentary porewater sources contributed 4–9% of DBC to Antarctic Bottom Water in Prydz Bay[38], and sediment resuspension could also contribute to the Bussol' Strait DBC inventory[45]. In our study region, the contribution of benthic inputs, including SGD, appears to be negligible for DBC concentrations, since DBC concentrations were independent of ^{228}Ra activities, while DOC concentrations, which are influenced mainly by benthic inputs, showed significant correlations with ^{228}Ra[40]. We also exclude potential contributions from hydrothermal vents[20] and autochthonous DBC production, as no such activities have been reported in this region. All these unknown sources and remineralization are likely reflected in the scatter of the

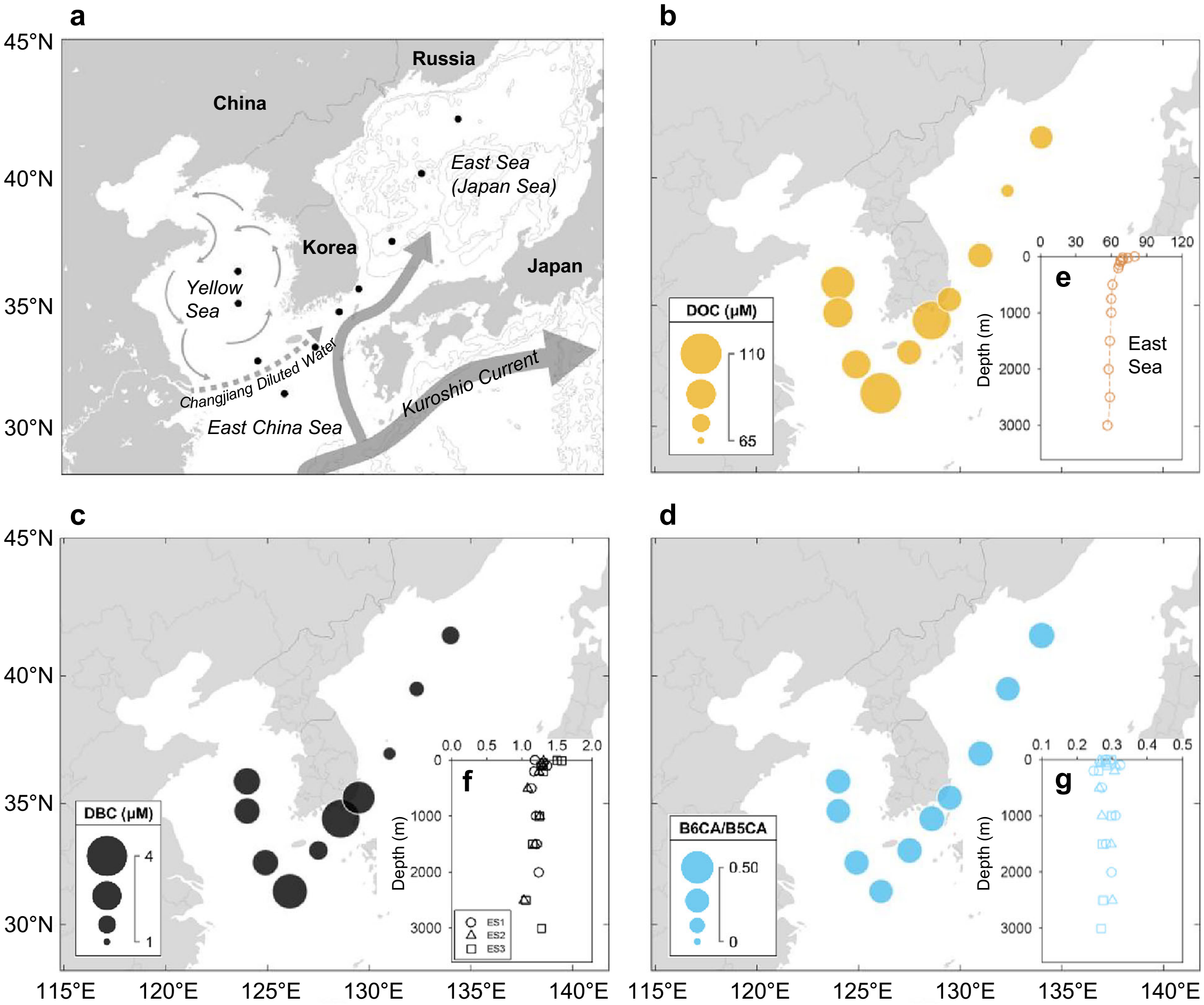

Fig. 1 | Distributions of dissolved organic carbon (DOC) and dissolved black carbon (DBC) in the northwestern Pacific marginal seas. a Map of the sampling stations, bottom topography, and simple schematic patterns of surface currents in the northwestern Pacific marginal seas. Distributions of **b** DOC concentrations, **c** DBC concentrations, and **d** the ratios of benzenehexacarboxylic acid to benzenepentacarboxylic acid (B6CA/B5CA) in the surface waters. Depth profiles of **e** DOC concentrations, **f** DBC concentrations, and **g** B6CA/B5CA ratios in the East Sea (Japan Sea). In panels f and g, open circles, triangles, and squares represent stations ES1, ES2, and ES3, respectively. DOC data for the vertical profile were obtained from Kim et al.[55].

salinity–DBC plot and are therefore insignificant in this semi-enclosed system.

The B6CA/B5CA ratio (average = 0.28 ± 0.02; $R^2 = 0.07$, $p = 0.15$, $n = 31$), an indicator of molecular condensation of DBC[16], was remarkably uniform across the study region, including the deep East Sea (Figs. 1g and 2c). Because the highly condensed fraction of DBC is preferentially removed by photodegradation[16], prolonged UV exposure in shelf waters would be expected to decrease this ratio. However, the uniform ratio observed across the entire marginal sea system, including the Yellow Sea (water residence time: ~5 years) and the East Sea (water residence time: ~100 years), suggests limited photochemical alteration of DBC in this system.

To roughly constrain the DBC budget in the northwestern Pacific marginal seas, we performed a first-order estimation based on available data (Fig. 3). Details of the calculation procedures, assumptions, and data sources are provided in the Methods. Here, the excess flux refers to values exceeding the Kuroshio background concentration. The riverine DBC flux (118 ± 12 Gg C yr^{-1}) dominates the external contributions, and atmospheric deposition (3–21 Gg C yr^{-1} to the ECS shelf and 4–28 Gg C yr^{-1} to the East Sea) was identified as a secondary source. As mentioned above, other unknown sources and sinks were assumed to play only a minor role in the region and therefore were not considered in our budget estimate. The transfer fluxes of excess DBC to the East Sea surface and deep layers were estimated to be 96 ± 15 Gg C yr^{-1} and 66 ± 4 Gg C yr^{-1}, respectively. Accordingly, the DBC concentration in the Kuroshio Current (0.81 μM)[24] increased to 2.49 μM in the Yellow Sea. It was diluted to 1.32 μM in the East Sea surface waters and 1.19 μM in the deep layer. The export of DBC to the deep sea, based on the deep ocean mass balance, may be decoupled from the modern input fluxes as the production of DBC on land has changed over the ~100-year residence of deep water in the East Sea[46]. Despite large uncertainties in these estimates, the overall budget appears to be roughly balanced.

Although the observed conservative behavior of DBC across the northwestern Pacific marginal seas suggests no significant removal during transport, our data show that DBC concentrations in the deep East Sea are ~10% lower than those in the surface (Fig. 3). This vertical gradient, despite the expected homogeneity under conservative behavior, implies either removal in the deep water or increased inputs of DBC over the past 100 years. Although we cannot rule out the contribution of particle scavenging to the 10% decrease, the trend is more likely due to the time lags between historical inputs recorded in deep waters and enhanced modern inputs. Global BC emissions have substantially increased over the past century, rising from 2.4 Tg yr^{-1} in 1700 to 3.7 Tg yr^{-1} in 1900, and further to 7.9 Tg yr^{-1} in 2021[46]. Although

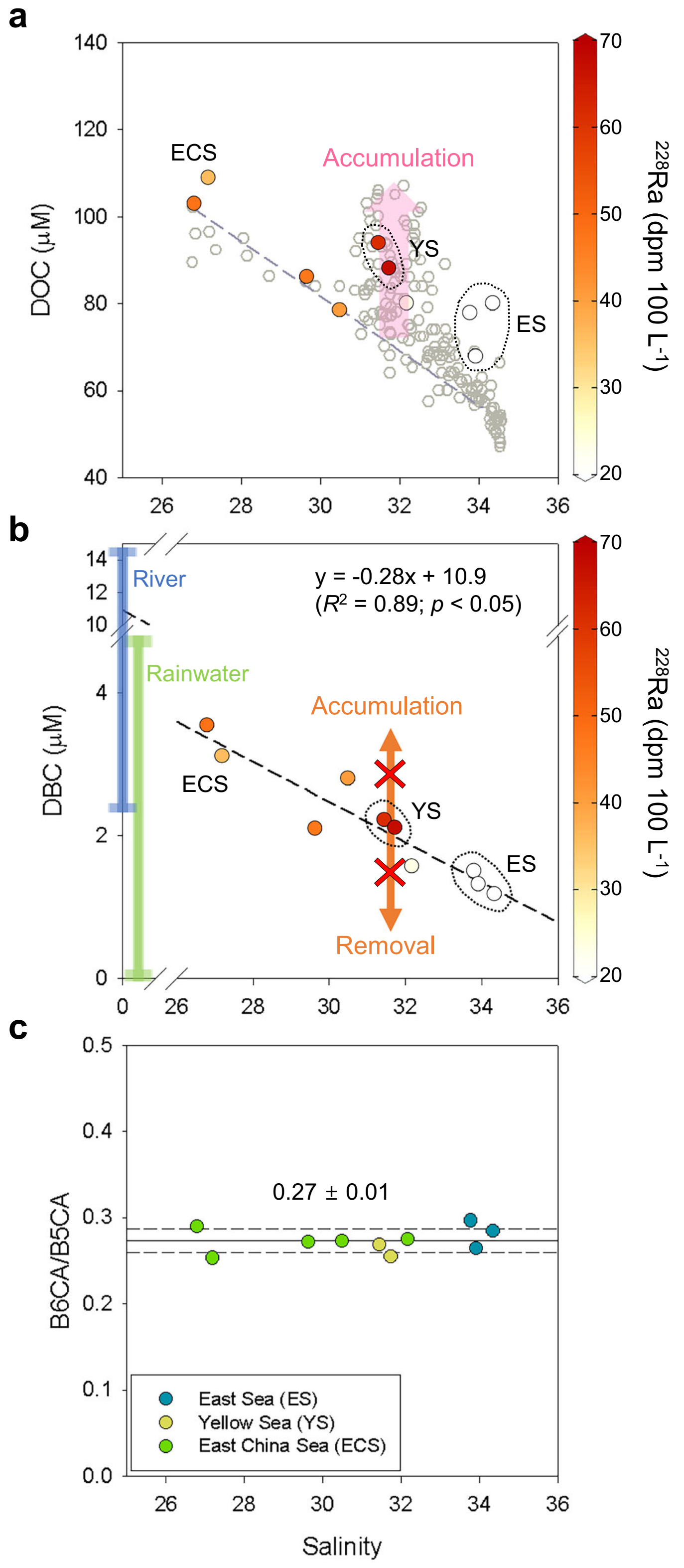

Fig. 2 | Relationships between salinity and dissolved organic carbon (DOC) and dissolved black carbon (DBC) concentrations, and the ratios of benzenehexacarboxylic acid to benzenepentacarboxylic acid (B6CA/B5CA) in the northwestern Pacific marginal seas. a Relationship between salinity and DOC concentrations in the surface waters of the northwestern Pacific marginal seas, including the East China Sea (ECS), the Yellow Sea (YS), and the East Sea (ES). Reference data from Han et al.[56] for ECS and YS are shown as gray open circles. The gray dashed line represents the endmember mixing line between the Changjiang diluted water and the Kuroshio Current water. The color gradient indicates ^{228}Ra activity, and ^{228}Ra activities in the ES were obtained from Cho et al.[57], and those for the ECS and YS were obtained from Park et al.[41]. **b** Relationship between salinity and DBC concentrations. The black dashed line denotes the linear regression ($R^2 = 0.89$, $p < 0.001$, $n = 10$). **c** Relationship between salinity and the B6CA/B5CA ratios. The solid line and dashed line represent the average and standard deviation of the B6CA/B5CA ratios, respectively.

not all emitted BC is transported to the ocean or transformed into DBC, this temporal trend may have contributed to elevated oceanic DBC inputs via both riverine and atmospheric pathways, leading to the observed depth gradient, as deep waters still reflect lower past inputs. If we assume that about half of the DBC concentration in the modern atmosphere is from anthropogenic activities[46] and that the DBC concentration in precipitation is fivefold lower than that of river water[43], the contribution of anthropogenic DBC corresponds to ~10% of that in river water. Similarly, about 5–18% of DBC in the catchment water of the Paraíba do Sul River in Southeast Brazil originated from aerosols[47]. We do not have enough information on historical changes in other factors, such as wildfire events, land use, and fuelwood, that may control riverine DBC inputs in this region. However, the source region of this study has been greatly influenced by human activities during the past century. It is therefore plausible that anthropogenic enhancement of atmospheric and riverine inputs has increased DBC concentrations in the surface layer by ~10%.

On a global scale, the average ^{14}C age of oceanic DBC in the deep ocean (23,000 ± 3000 ^{14}C years)[10] is much older than the ~800 years estimated from global riverine fluxes of DBC and the oceanic standing stock[10], which has led to suggestions that DBC is removed before reaching the deep ocean. Considering that this marginal-sea system is characterized by large terrestrial inputs and removals, prolonged exposure to sunlight in surface waters, and long residence time (~100 years) in the deep sea, our observational results, including the remarkably uniform B6CA/B5CA ratios across regions, demonstrate that DBC is highly conservative not only in the northwestern Pacific marginal seas but also in the other major source regions. Furthermore, the conservative behavior of DBC was also observed in the Southern Ocean[26]. Thus, the DBC concentration in the global deep ocean could be significantly lower than that expected from river inputs due to enhanced modern inputs rather than removal processes in source regions. Considering the fact that the average age of the North Pacific water is ~1500 years[48], human activities over the past few centuries could have substantially increased DBC inputs, which were likely reflected in riverine fluxes but not yet in the aged deep ocean. For example, the large-scale destruction of Brazil's Atlantic forest was estimated to have produced 2.8–7.2 million tons of BC within ~120 years[49]. In addition to enhanced modern inputs of DBC, preferential degradation of younger riverine DBC could also have contributed to the old ^{14}C ages observed in the deep ocean, since the ages and degradation of DBC are variable in the ocean[50]. Such fractionations by degradation or historical changes in the contribution of C4 plants could also result in ~6‰ enrichment in stable carbon isotope values[19].

By demonstrating the main source inputs and conservative behaviors of DBC in this unique marginal sea system, our results provide a basis for refining global cycles of refractory DOC as well as DBC in the ocean. Our findings should be further validated in other oceanographic settings. We also suggest that further studies are needed to elucidate historical changes in riverine DBC inputs and age-fractionated degradation of DBC in the global ocean.

Methods

Sample collection

Sampling was conducted in August 2020 (ECS1–5 and YS1–2), February 2018 (ES1), December 2018 (ES2), and November 2019 (ES3) aboard the

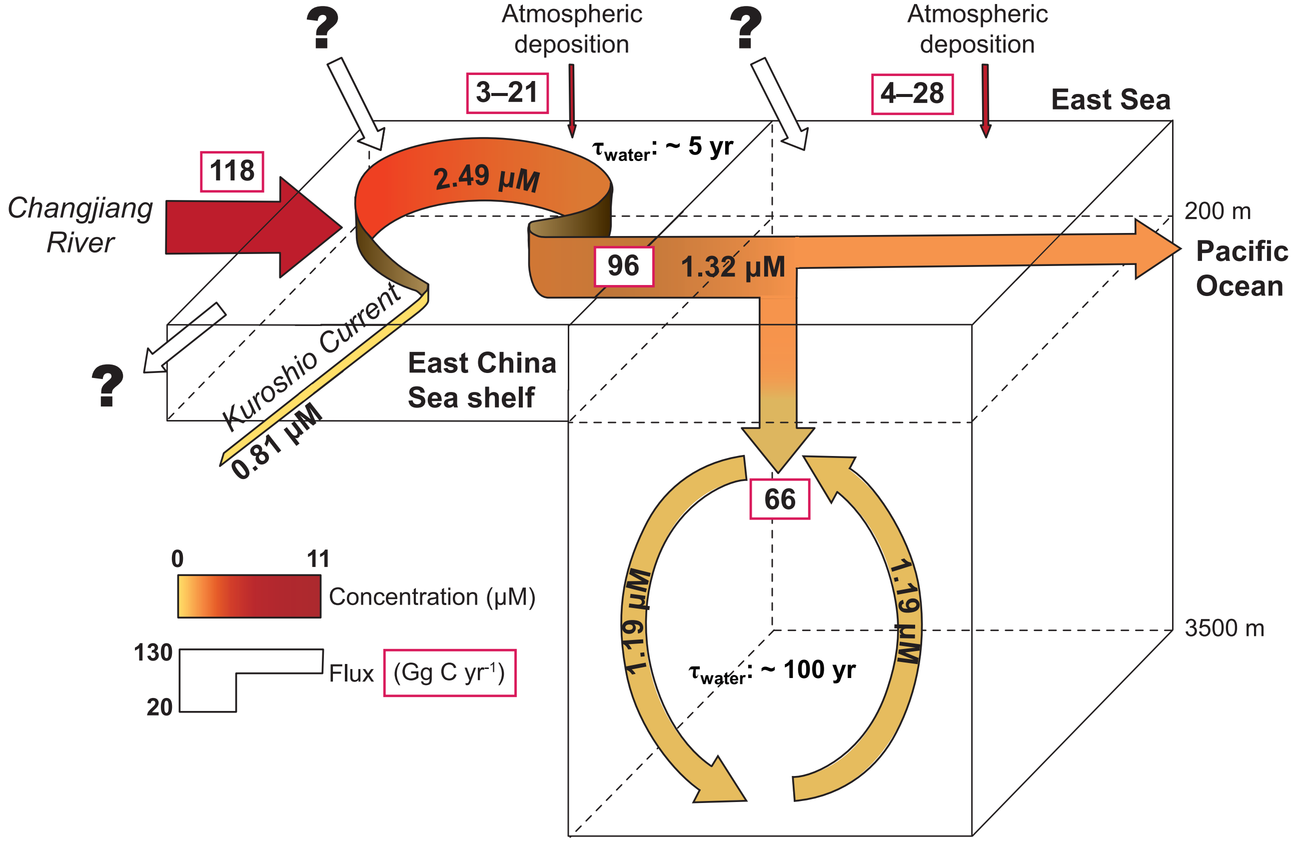

Fig. 3 | Schematic of the dissolved black carbon (DBC) in the northwestern Pacific marginal seas. DBC transport and distribution in the East China Sea shelf and the East Sea are illustrated. Arrows represent DBC fluxes, arrow color indicates DBC concentrations (μM), and arrow width is proportional to the magnitude of flux (Gg C yr^{-1}).

R/V *Onnuri* of the Korea Institute of Ocean Science and Technology (KIOST), the R/V *Isabu* of KIOST, the R/V *Isabu* of KIOST, and the R/V *Akademik Oparin* of the Pacific Oceanological Institute (POI, Russia), respectively (Fig. 1a). All seawater samples were collected using 10 L Niskin bottles mounted to the CTD rosette from ten sampling stations. In the ECS and the Yellow Sea, sampling was conducted for surface water only. In the East Sea, sampling was conducted for the whole water column, with depths ranging from surface to 2000–3000 m depending on the station. Samples for DOC analysis were filtered onboard through precombusted (450 °C, 5 h) GF/F filters (0.7 μm pore size; Whatman) and acidified to pH 2 with HCl. Samples for DBC analysis were filtered onboard through acid-cleaned polyethersulfone capsule filters (0.45 μm pore size; AcroPak-200, Pall) and acidified to pH 2 with HNO_3.

DOC analysis

DOC concentrations were measured using a high-temperature catalytic oxidation method with a total organic carbon analyzer (TOC-L, Shimadzu) and quantified based on a calibration curve made with a potassium hydrogen phthalate stock solution. Measurement accuracy and reproducibility were assessed using deep-sea reference samples provided by the University of Miami (D. Hansell), with all measurements agreeing within 5% of reported values.

DBC analysis

Seawater samples (~1 L) for DBC analysis were concentrated using a solid-phase extraction (SPE) method with PPL resin (1 g, Agilent), following the procedure described by Dittmar et al.[51]. The extract was eluted from the resin using HPLC-grade methanol (~9 mL) and dried at 50 °C. The average recovery of DOC by SPE was 57 ± 7%.

DBC concentrations were determined using the benzenepolycarboxylic acid (BPCA) method described by Dittmar[52]. Concentrated HNO_3 was added to the samples in glass ampoules, which were then thermo-chemically oxidized at 170 °C for 8 h. The acid was evaporated using a speed vacuum at 60 °C for 4 h, and the oxidation products were subsequently re-dissolved in phosphate buffer solution. BPCAs were determined using ultrahigh performance liquid chromatography with a photo-diode array detector (UPLC-PDA, Acquity, Waters). DBC concentrations were calculated from BPCA concentrations using the following equation: $[DBC] = 0.0891 \times ([B5CA + B6CA])^{0.9175}$[16,53].

Previously published data were obtained from samples acidified with HCl instead of HNO_3. To assess the potential influence of acid type on the BPCA method, a methodological test was conducted using water from the Wadden Sea near Spiekeroog, North Sea. No significant differences were observed in any BPCA-related parameters between HCl- and HNO_3-acidified samples (DBC: $p = 0.655$; B5CA: $p = 0.543$; B6CA: $p = 0.938$; DBC yield: $p = 0.566$; B5CA yield: $p = 0.475$; B6CA yield: $p = 0.869$).

Statistical analysis

The *t*-test or Mann–Whitney *U* test was conducted to evaluate the significance of the variations in the concentrations of DOC and DBC, and B6CA/B5CA ratios between different depth layers of seawater (surface vs. deep layer). The normality and homogeneity of data were analysed using the Shapiro–Wilk test and Levene's test, respectively. All statistical analyses were conducted using SPSS 29 software for Windows (IBM Corporation). The significance level was set at $p < 0.05$ for all performed statistical tests.

Estimation of the DBC budget

To constrain the DBC budget in the northwestern Pacific marginal seas, we performed a first-order estimation under the assumption of steady-state conditions (Fig. 3). The riverine endmember concentration of DBC was obtained by extrapolating the DBC–salinity relationship to zero salinity, which yielded 10.9 ± 1.1 μM (95% confidence level by regression analysis) and was taken to represent the Changjiang River input. The corresponding riverine DBC flux was estimated to be 118 ± 12 Gg C yr^{-1} from this endmember concentration and the annual river discharge of 899 km^3 yr^{-1}[54]. Since this river endmember is estimated from the extrapolation of samples collected across different locations, seasons, and water ages, contributions from other rivers may have been included, although only the discharge volume of the Changjiang River was used.

Atmospheric deposition to the ECS shelf was estimated using literature-based deposition rates (0.34 mmol m^{-2} yr^{-1} in the offshore and 2.3 mmol m^{-2} yr^{-1} in the nearshore area)[24] and the surface area of the ECS shelf, yielding 3–21 Gg C yr^{-1}. For the East Sea, atmospheric deposition was estimated as 4–28 Gg C yr^{-1} using its surface area and the same deposition rates. Since literature data are rare, we allow extreme ranges that may include temporal and seasonal variations in DBC inputs.

The excess DBC (i.e., the flux in excess of the Kuroshio background concentration) exported from the ECS shelf to the East Sea was estimated from the difference between shelf DBC concentrations and the Kuroshio value (0.81 μM)[24], multiplied by the water transport of 0.11 Sv, giving 96 ± 15 Gg C yr^{-1}. The uncertainty of the shelf DBC excess flux was evaluated by error propagation from the standard error of shelf concentrations ($n = 2$), assuming that the Kuroshio endmember was constant. The input flux of DBC to the deep layer (200–3000 m) of the East Sea was estimated to be 66 ± 4 Gg C yr^{-1}, based on the excess DBC inventory divided by the water residence time (100 years). The uncertainty of this flux was propagated from the standard error of deep-water concentrations ($n = 15$).

Data availability

The data generated in this study have been deposited in the figshare repository (https://doi.org/10.6084/m9.figshare.30145351) and are also provided in the Supplementary Information.

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Author contributions

J.P., M.R., H.W., and G.K. contributed to the design of the research. H.H. and H.S. contributed to the field observations and sample collection. J.P. and M.R. performed DBC analyses under the supervision of H.W. and T.D. J.P., M.R., H.W., and G.K. carried out the data analyses, with additional contributions to data interpretation from I.-S.H. and J.-S.L. All authors contributed to the writing and revision of the manuscript.

Competing interests

The authors declare no competing interests.

Additional information

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