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Article

Global patterns and drivers of dissolved organic matter across Earth systems: insights from H/C and O/C ratios

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ABSTRACT

Dissolved organic matter (DOM) is ubiquitous and contains a complex pool of thousands of distinct molecules, and their chemical characteristics help us inform the fate of global carbon. However, a more holistic perspective of the molecular characteristics of DOM and underlying mechanisms across Earth systems and climates remains under study. Here, we present a comprehensive analysis of the molecular characteristics of DOM using two abundance-weighted average indices, i.e., H/C and O/C ratios, by compiling 2,995 samples from 317 studies covering waters, land, plant, petroleum, and atmosphere systems and climatic regions from the tropics to tundra. H/C ratios are lower on average in waters (H/C = 1.15 ± 0.005) and land (H/C = 1.20 ± 0.010) than in the other systems, while their O/C ratios rank between plant and atmosphere systems. In the waters and land systems, the H/C ratios of DOM vary from the highest to the lowest in the habitats of the land-to-ocean continuum generally as snow > glacier > marine ≥ freshwater/soil > groundwater. The H/C ratios show predictably U-shaped patterns along latitudinal gradients, indicating the lowest abundance of more hydrogen-saturated molecules at mid-latitudes of approximately 40°-50° in river water, lake water, and forest soil. The two ratios are primarily controlled by environmental factors such as pH, dissolved oxygen, and carbon and nitrogen contents. We further unveil additional and considerable links between the ratios and the extremes of climatic factors such as precipitation of warmest quarter and maximum temperature of warmest month. Our synthesis provides molecular-level perspectives to characterize the global distribution and underlying drivers of DOM, which is complementary for our understanding of global carbon cycle processes under future global change.

1. Introduction

Dissolved organic matter (DOM) is an essential component of the Earth's biogeochemical cycles in determining carbon sources or sinks and is ubiquitous within and across Earth systems [1–3]. Terrestrial soil, inland waters, and the ocean are key carbon reservoirs, exerting control over greenhouse gasses in the atmosphere and influencing Earth's climate, as they not only transport and process but also bury large amounts of organic carbon [2,4–6]. For example, global carbon sequestration in soil, inland waters, and ocean are estimated at ~ 0.9, 0.15, and 0.2 Pg per year, respectively [7,8]. The interconnectedness of these systems, as seen in the emerging global carbon cycle perspective, results in the exchange of dissolved carbon and nutrients. This, in turn, impacts the fate of DOM, such as affecting decomposition and production rates [5,9]. The global carbon cycle processes could be effectively informed from the molecular-level perspectives of DOM characteristics (i.e., molecular traits) [3,10–12]. A better understanding of the regulatory mechanisms of the global variation in DOM traits resulting from the spatial heterogeneity of climatic and environmental variables is important for estimating the responses of carbon cycle processes to environmental changes. Incorporation of the previously overlooked drivers into predictive models (e.g., Earth system models) is needed to reduce the uncertainty of estimation [13,14]. Therefore, it is crucial to develop a more holistic perspective of the distribution and underlying drivers of DOM molecular traits across Earth systems and multigradient environments, which ultimately helps inform modeling for predicting future global carbon cycle processes.

Organic matter chemistry is a complex pool of thousands of distinct molecules, each with unique molecular traits, primarily characterized by the hydrogen-to-carbon ratio (H/C) and oxygen-to-carbon ratio (O/C)

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[15,16]. The H/C ratio is relevant to biogeochemical reactions of hydrogenation or dehydrogenation, reflecting the degree of hydrogen saturation. A higher H/C ratio reflects a higher degree of hydrogen saturation. The H/C ratio can also be applied to indicate the capacity for a molecule to be degraded [17,18]. The O/C ratio is relevant to chemical reactions of oxidation or reduction. A higher O/C ratio reflects a higher degree of oxygenation and more oxygen-containing functional groups, such as carboxyl or hydroxyl groups [19]. These two dimensions of traits could be constrained by microbes and environmental conditions such as nutrients, temperature, and sunlight, and further inform the transformation of organic matter [20]. For example, environmental conditions such as low oxygen availability can enrich compounds with lower oxygenation and make organic matter degradation thermodynamically unfeasible [21,22]. Climatic factors such as the mean annual temperature can also affect DOM characteristics, with more hydrogen-saturated compounds enriched in higher temperature conditions [19]. Despite the well-known importance of the average state of the climatic variables, there are few investigations considering the influences of extremes or variability in climatic variables on DOM composition.

Here, we compiled compositional-level H/C and O/C ratios of DOM from 3,558 samples derived from 317 studies spanning diverse systems and climates worldwide (Fig. 1, Fig. S1, and Table S1). The datasets included waters, land, plant, petroleum, and atmosphere systems, covering the climatic regions from the tropics to the tundra (Fig. 1, Fig. S1, and Table S1). There were 2,876 samples (80.8%) from waters and land covering the habitats of the land-to-ocean continuum, e.g., soil, peatland, glacier, pond, reservoir, lake, river, and ocean. The availability of such large datasets benefits from the recent advances in ultrahigh-resolution Fourier Transform Ion Cyclotron Resonance Mass Spectrometry (FT-ICR MS) [15,16]. FT-ICR MS has been applied to numerous natural organic matter such as terrestrial, aquatic and marine DOM, microbial-derived DOM, and petroleum-derived materials, and to further determine chemical characteristics (i.e., molecular traits) as a function of (bio)geochemical or anthropogenic processes [12,23,24]. Compositional-level DOM traits reduce the complex mass spectrum data for the individual peaks to abundance-weighted average indices [15]. This is beneficial for an intersample comparison by incorporating climatic and environmental factors and has already been well applied in

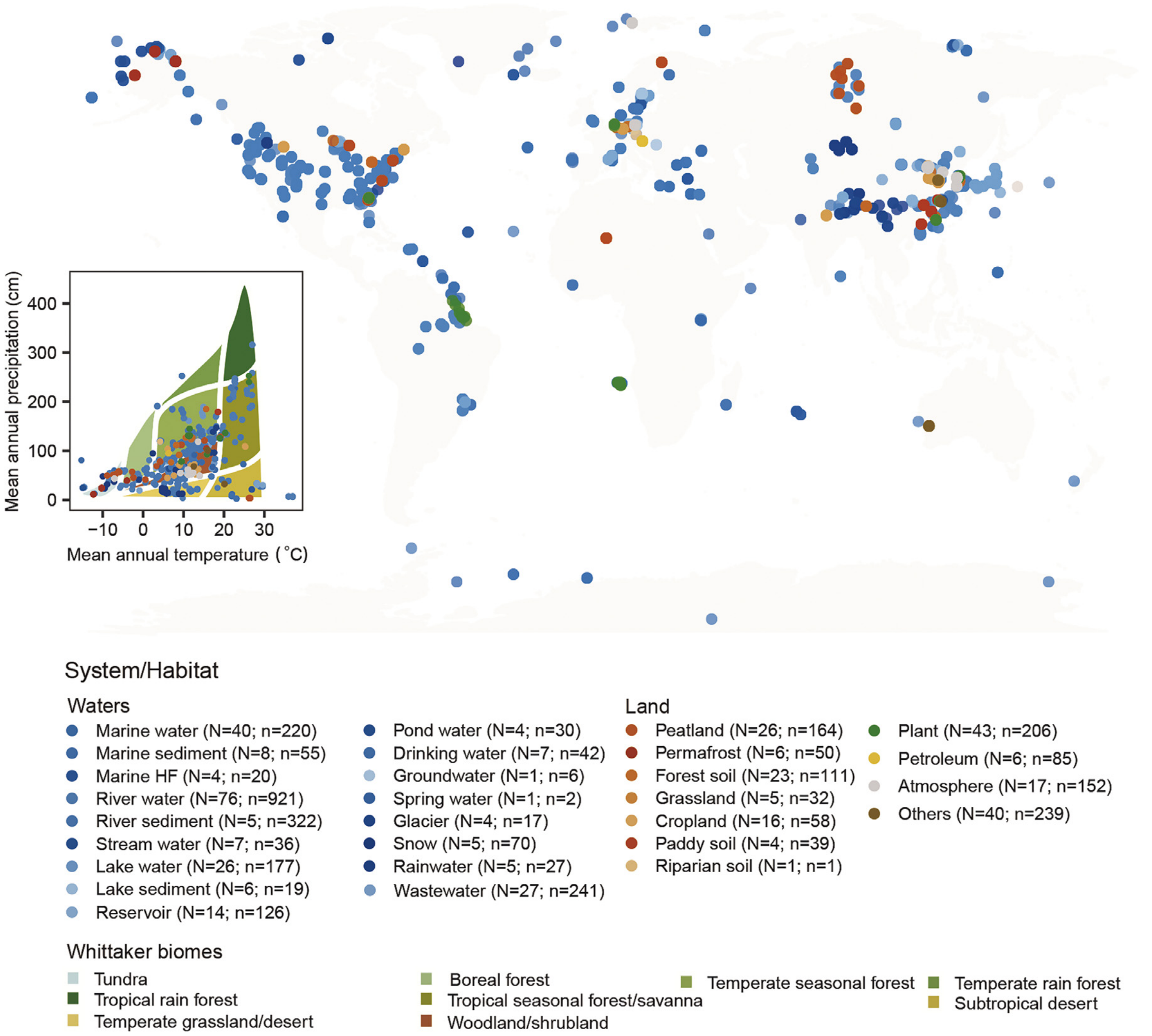

Fig. 1. Map of samples included in the compiled dataset of H/C and O/C ratios of DOM. We obtained a total of 3,558 samples from 317 studies that measured H/C and O/C ratios across Earth systems, including waters, land, plant, petroleum, and atmosphere, prior to June 2022. The geographic locations of samples across different habitats are shown with colored dots. Numbers of studies (N) and samples (n) per system are given in the parentheses. The inset figure shows the distribution of samples across the gradients of temperature and precipitation. Polygons depict Whittaker's biomes [67] according to mean annual temperature (°C) and mean annual precipitation (mm yr^{-1}): (1) tropical rainforest; (2) tropical seasonal rainforest/savanna; (3) subtropical desert; (4) temperate rainforest; (5) temperate seasonal forest; (6) woodland/shrubland; (7) temperate grassland/desert; (8) boreal forest; and (9) tundra. The colored dots indicate the samples across different habitats. The full publication list for the global synthesis is shown in Supporting Information Table S1.

DOM studies [1,10,19]. We aimed to provide a comprehensive survey on molecular-level perspectives of DOM characteristics at a global scale. Specifically, the synthesis explores the distribution patterns of H/C and O/C ratios of DOM across Earth systems and along latitudinal gradients and elucidates the roles of climatic and environmental variables in driving these traits. Such global patterns and drivers for DOM via a meta-synthesis study could be more important when estimating the effects of global environmental change on carbon cycle processes given the limited scopes of individual studies.

2. Materials and methods

2.1. Data collection

We systematically searched all peer-reviewed publications that were published prior to June 2022, which investigated the molecular traits (i.e., H/C and O/C ratios) of DOM measured by FT-ICR MS using the Web of Science (Core Collection; http://www.webofknowledge.com) and Google Scholar (http://scholar.google.com) via the search terms: "organic matter AND FT-ICR MS AND van Krevelen". The molecular traits of thousands of molecular formulae (hereafter referred to as "molecules") for each sample's FT-ICR MS spectrum were evaluated on van Krevelen diagrams on the basis of their molar H/C ratios (y axis) and molar O/C ratios (x axis) [15]. The van Krevelen diagrams enable the comparison of molecular properties of organic matter and the ability to assign molecules to major biochemical categories, which include amino sugar-, lipid-, protein-, lignin-, carbohydrate-, tannin-, and condensed aromatic-like compounds. However, it should be noted that we here used van Krevelen diagrams to visualize the H/C and O/C ratios at the compositional level, and the sample points in the diagrams do not intend to assign the samples to these biochemical categories.

We employed the following criteria to select the studies. (1) They had raw mass spectrometry data, from which the compositional-level H/C and O/C ratios could be calculated. (2) They had compositional-level H/C and O/C ratios, which are weighted means of formula-based H/C and O/C ratios in a given sample, which are calculated as the sum of the H/C (or O/C) ratio for each molecule and its relative intensity divided by the sum of all intensities [1,10]. (3) They focused on the DOM extracted from natural and engineered environments, rather than manipulated experiments. In total, the H/C and O/C ratios of 3,558 samples from 317 studies met these criteria (Table S1).

To minimize the challenges in data comparison and interpretation across studies with different instrument types and settings [25,26], we employed the following criteria to further subset the data: (1) DOM trait datasets obtained by FT-ICR MS were retained, but not by other instrument types such as Orbitrap MS. (2) Negative ESI mode was retained for the following statistical analyses, as it is most frequently documented in the literature by comprising 88.9% of the total datasets and is the most suitable ionization method for the analysis of natural DOM. (3) We focused on the compositional-level H/C and O/C ratios calculated based on all molecules in a given sample, rather than the samples with only subsets of molecules. In total, there were H/C and O/C ratios of 2,995 samples from 270 studies using (-)ESI-FT-ICR MS for the robust data comparison among various systems and habitats.

The collected dataset included various Earth systems, such as waters, land, plant, petroleum, and atmosphere, covering climatic regions from the tropics to tundra (Fig. 1, Figs. S1, S2, and Table S2). We further binned the dataset of each system into fine habitats (Figs. S3, S4, Table S3). Specifically, the waters include habitats of marine water, marine sediment, marine hydrothermal fluid, lake water, lake sediment, reservoir, pond, river water, river sediment, stream, drink water, groundwater, spring, glacier, snow, rainwater, and wastewater. The land includes habitats of peatland, permafrost, forest soil, grassland, cropland, paddy soil, riparian soil, and coastal soil. The plant includes habitats of phycophyta, herbage, arbor, and shrub. The atmosphere includes habitats of aerosol, particulate matter (PM) 2.5, and PM 10. The glacier is mainly derived from marine ice and lake ice. There were several habitats categorized as "Others", including virus, melanin, murchison, mineral, coal, biochar, and manure. Waters and land systems were discussed in more detail than plant, petroleum, and atmosphere systems, as more sufficient data derived from these two systems and their finely categorized habitats were available in the literature. It should be noted that we also included rarely reported systems such as plant, petroleum, and atmosphere systems, as this synthesis was aimed to provide an overview for comparing DOM traits derived from as many Earth systems as possible.

In addition to the molecular traits of H/C and O/C ratios, we further collected climatic and environmental variables for each sample via the WorldClim dataset (https://www.worldclim.org) and the original studies (Table S1). A total of 15 environmental variables were collected, including salinity, temperature, pH, conductivity, and the concentrations of dissolved oxygen (DO), total organic carbon (TOC), total nitrogen (TN), total dissolved nitrogen (TDN), dissolved organic carbon (DOC), ammonium (NH_4^+), nitrate (NO_3^-), nitrite (NO_2^-), phosphate (PO_4^{3-}), iron (Fe), and manganese (Mn). In addition, climatic variables were derived using latitude, longitude and digital elevation data with a spatial resolution of 0.5°. The gridded data were obtained from the WorldClim dataset for the 19 bioclimatic variables [27,28], including annual mean temperature (BIO1), mean diurnal range (BIO2), isothermality (BIO3), temperature seasonality (BIO4), maximum temperature of warmest month (BIO5), minimum temperature of coldest month (BIO6), temperature annual range (BIO7), mean temperature of wettest quarter (BIO8), mean temperature of driest quarter (BIO9), mean temperature of warmest quarter (BIO10), mean temperature of coldest quarter (BIO11), annual precipitation (BIO12), precipitation of wettest month (BIO13), precipitation of driest month (BIO14), precipitation seasonality (BIO15), precipitation of wettest quarter (BIO16), precipitation of driest quarter (BIO17), precipitation of warmest quarter (BIO18), and precipitation of coldest quarter (BIO19).

2.2. Statistical analysis

The significance of differences in H/C or O/C ratios between Earth systems was determined using a Kruskal-Wallis test. Pairwise comparison was performed for the magnitude of variances of H/C or O/C ratios between habitats using the Wilcoxon test. These analyses were performed using the R package stats V4.1.3.

We further explored the distribution patterns of compositional-level H/C or O/C ratios along latitudinal gradients and the influences of explanatory variables on these two ratios. The explanatory variables included 19 bioclimatic and 15 collected environmental variables. It should be noted that although DOM molecular traits are also dependent on microbes and sunlight, we here focused on climatic vs. environmental constraints for the following reasons: (1) there were important roles of climatic and environmental variables documented in previous literature, such as Roth *et al.*, (2019) [1] and Hu *et al.*, (2022) [10]; (2) microbial data are not always available along with DOM mass spectral data in the same literature, and thus, the influences of microbes and sunlight on DOM could not be well quantified in our meta-analysis study. For better statistical power, we performed the analyses with a sample size over 30 for the waters or land, or each of their habitats.

The latitudinal patterns of the H/C and O/C ratios were fitted using generalized additive models [29]. The influences of climatic and environmental variables on H/C and O/C ratios were evaluated by linear mixed-effects models [30]. In each model, we modeled H/C or O/C ratios in every Earth system (that is, waters or land) as a function of a climatic or environmental variable and used studies and habitats as random effects. The omnibus test was used to evaluate model significance, and the conditionally explained heterogeneity represented the influence of each explanatory variable on the H/C or O/C ratios accounting for the random effects [31]. To minimize the potential biases of data discrepancies across various instruments or laboratories, we specified random effects in our model, which are able to factor out the idiosyncrasies of our samples and obtain a more general estimate of the fixed effects of interest [32]. We further examined the influences of each of these explanatory variables on the H/C or O/C ratios in each habitat of waters and land, in which we used the identity of data-source studies as random effects. The analyses of linear mixed-effects models were performed by using lmer function in the R package lme4 V1.1.28. This approach enabled us to obtain reliable results of the latitudinal patterns of H/C or O/C ratios and the influences of climatic and environmental variables on the ratios. Further partitioning analysis in linear mixed-effects models provided an estimate of the total contribution of a fixed effect of each climatic variable to the overall prediction of the H/C or O/C ratio. We selected climatic variables for partitioning analyses by dereplicating strongly correlated variables by a threshold of Pearson correlation over 0.8. Partitioning analysis was performed with R package partR2 V0.9.1 [33].

3. Results and discussion

3.1. Variation in DOM traits across Earth systems

The molecular traits of DOM, measured by H/C and O/C ratios at the compositional level (hereafter, H/C and O/C ratios), were highly divergent across Earth systems, such as in waters, land, plant, petroleum, and atmosphere systems (Fig. 2a, Fig. S2, Table S2). The H/C and O/C ratios varied from 0.22 to 2.14 and 0.01 to 1.04, with mean values of 1.17 and 0.41, respectively, in all systems (Fig. 2a, Table S2). H/C ratios were lower than 1.5 in 92.0% of samples, indicating that DOM generally contained a high abundance of recalcitrant (i.e., less hydrogen saturated) molecules in each system [18].

Among these systems, atmosphere samples showed the highest mean values for H/C (mean $\pm$ s.e = 1.47 $\pm$ 0.030) and O/C ratios (0.45 $\pm$ 0.017), indicating a higher abundance of more hydrogen-saturated molecules and more abundant oxygen-containing functional groups [15,19], respectively (Fig. 2b, Table S2). The atmosphere experiences rapid photochemical transformation and, therefore, indicates that DOM contains a higher abundance of more hydrogen-saturated molecules than other systems and thus the highest H/C mean value. In comparison, petroleum samples had relatively intermediate H/C (mean = 1.37 $\pm$ 0.034) and the lowest O/C (mean = 0.29 $\pm$ 0.020) ratios, while plant had similar H/C and O/C ratios to those in waters and land (Fig. 2, Table S2).

There were 2,140 and 401 samples for waters and land, comprising 71.5% and 13.4% of the collected datasets, respectively (Fig. 1, Table S2). The mean values of the O/C ratios were significantly ($P \leq 0.05$) lower in waters (0.40 $\pm$ 0.002) than in land (0.43 $\pm$ 0.005), and their H/C ratios showed a similar pattern, with mean values of 1.15 $\pm$ 0.005 and 1.20 $\pm$ 0.011, respectively (Fig. 2, Table S2). We recognized the overlapping nature between waters and land systems, where the mean values of H/C ratios were lower than those of the other systems and their O/C ratios ranked between plant and atmosphere (Fig. 2, Fig. S2). This indicates that DOM in waters and land systems contains a higher abundance of molecules with more recalcitrant and relatively intermediate oxygenation than the other systems.

3.2. Variation in DOM traits across habitats of waters and land

When considering individual habitats of waters or land, we also found some distinct variations in H/C or O/C ratios between habitats (Fig. 3, Fig. S4, Table S3), and the significance tests are shown in Fig. S5. For the waters, H/C ratios were over 1.4 in 12.9% of samples and showed the significantly highest mean values in snow and rainwater ($P \leq 0.05$; Fig. 3, Fig. S5). Relatively high values of these two natural aquatic habitats were similar to those of the atmosphere system, which could be explained by their shared atmospheric source of DOM. Subsequently, the habitats such as stream, pond and spring had mean H/C ratios ranging from 1.29 to 1.31 (Fig. 3c). Similar H/C ratios were also observed in glacier and ocean, with mean values of 1.27 to 1.34 (Fig. 3c). These habitats had relatively higher mean values of H/C ratios over 1.2, while their O/C ratios ranged between 0.4 and 0.5 except for those in snow (Fig. 3c). Generally, the O/C ratios in each habitat showed significantly higher variation than the H/C ratios ($P \leq 0.05$; Fig. 4).

In contrast, river and drinking water showed lower means of H/C ratios ranging from 1.08 to 1.10, followed by groundwater, with the lowest value of 0.98 (Fig. 3c). We observed significantly lower values in these habitats than in most of the other aquatic habitats ($P \leq 0.05$; Fig. S5). Like lake, pond and stream, DOM characteristics in river are also influenced by terrestrial inputs [34]; however, lower H/C mean values are associated with lower carbon productivity and turnover rates due to flowing waters' short residence time [35]. As an engineered aquatic habitat, DOM in drinking water is characterized as the rapid microbial processing of labile DOM [36], leaving recalcitrant molecules behind and thus low H/C mean values. The lowest H/C ratios in groundwater indicate higher aromaticity than usual for those in aquatic and terrestrial surface environments, which is consistent with previous reports [37,38]. Notably, H/C ratios in lakes showed relatively large variation between sediment and water ($P \leq 0.05$) and were lower in the former, with mean values of 1.01 and 1.25, respectively (Fig. 3c, Fig. S5). The lower hydrogen saturation of DOM in lake sediment is associated with accumulated recalcitrant molecules, which could be likely due to higher microbial diversity and carbon metabolism and thus include residues of microorganisms and degradation products of organic matter [39,40]. For O/C ratios, the highest and lowest mean values of 0.51 and 0.31 were observed in drinking water and river sediment, respectively (Fig. 3c). The higher O/C ratio in drinking water is largely caused by the mixture of natural organic matter from the water source, chemical disinfection, and disinfection byproducts, which usually have aromatic structures [41,42].

For the land, the mean H/C ratios showed the highest and lowest values of 1.32 and 1.04–1.07 observed in coastal and cropland/riparian soils, respectively (Fig. 3c). Interestingly, H/C ratios were not significantly different ($P > 0.05$) between coastal soil and ocean, while those in cropland and riparian soil were similar to those in river and lake sediment (Fig. S5). This phenomenon agrees with the fact that there is a land–water continuum for the spatial dynamics of DOM to be transported from terrestrial soils to inland waters and from tidal wetlands to the ocean. In the context of future climate warming, the increased extreme rainfall intensity and frequency would enhance carbon and nutrient transport from soil to inland waters and to the ocean [43,44]. Consequently, this mixing of organic carbon sources of contrasting reactivity might result in changes in organic matter degradation through priming processes [45]. Compared to H/C ratios, O/C ratios showed significantly higher variation within each habitat for most land habitats ($P \leq 0.05$; Fig. 4). This suggests that the variations in DOM characteristics, especially O/C ratios, in each habitat should be constrained, such as by climate and environmental conditions resulting from their global spatial heterogeneity.

Collectively, DOM traits, particularly H/C ratios, showed clear variations across Earth systems and habitats, while O/C ratios had less clear variation, such as among habitats of waters and land, and showed fewer significant differences between pairwise habitats (Fig. S5). Specifically, H/C ratios were on average lower in waters and land than in other systems such as plant, petroleum and atmosphere. In these two systems, the H/C ratios of DOM varied from the highest to the lowest in the habitats of the land-to-ocean continuum generally as snow, rainwater > glacier > coastal soil, ocean, stream, pond, permafrost > lake water, reservoir, peatland, paddy soil, forest soil, grassland > river > lake sediment, riparian soil, cropland > groundwater. Based on a smaller number of observations, previous studies have also tried to compare the H/C ratios of DOM across a limited number of habitats. For example, H/C ratios were higher for glacier, followed by ocean and freshwater [18,46], higher in lake water relative to lake sediment [39], and higher in paddy soil

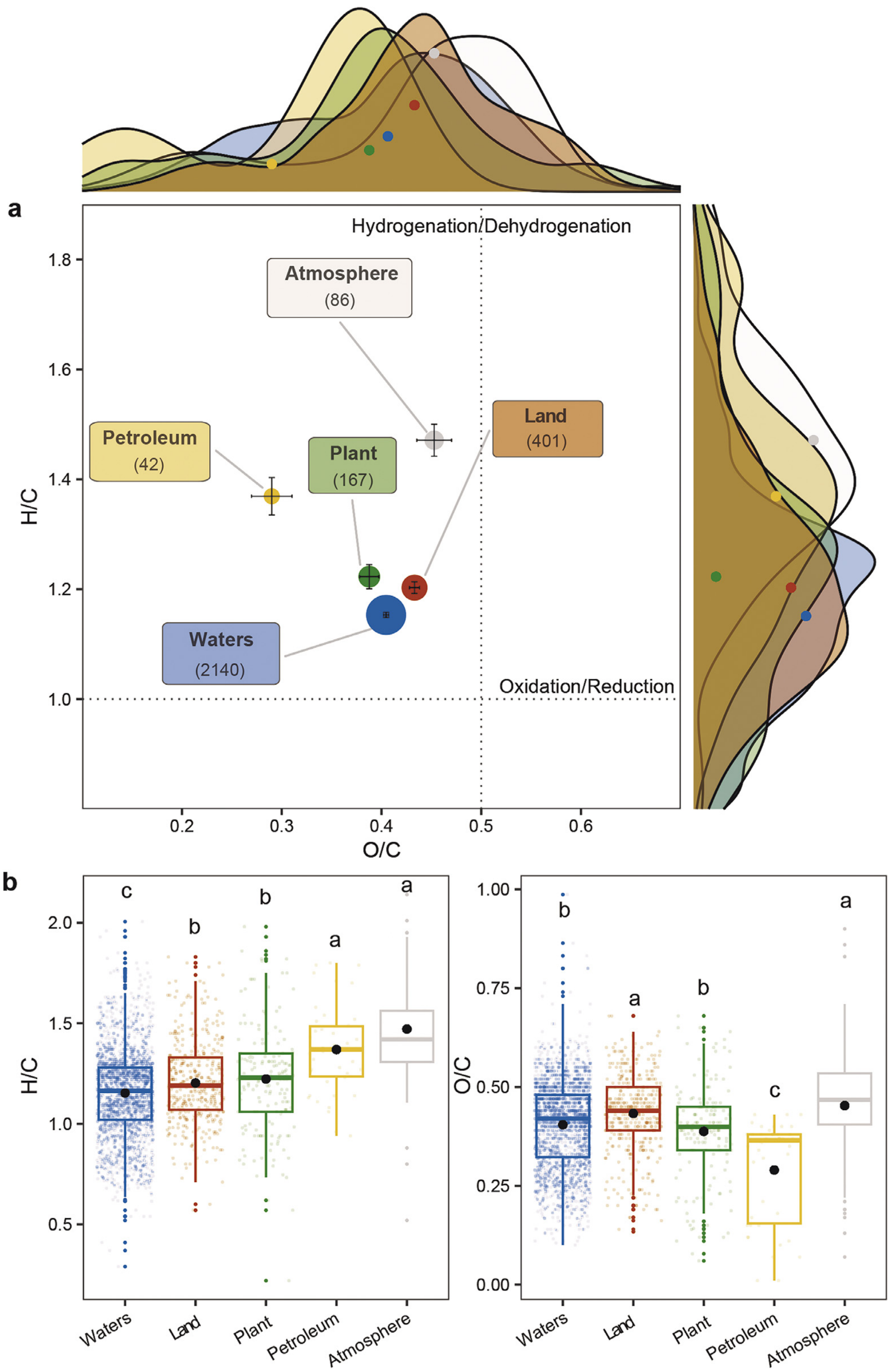

Fig. 2. Variation in H/C and O/C ratios of DOM across Earth systems. (a) Van Krevelen diagram shows the means of compositional-level H/C and O/C ratios measured by (-)ESI-FT-ICR MS in Earth systems, including waters, land, plant, petroleum, and atmosphere. The means ± s.e of H/C and O/C ratios are shown with colored dots, and the number of samples in each system is indicated by the dot size and in the parentheses. Black dashed lines represent the direction of change in H/C and O/C ratios for chemical reactions, including hydrogenation/dehydrogenation, and oxidation/reduction [12]. The marginal density plot shows the distribution of compositional-level H/C and O/C ratios in each system, and the small colored dots are their means. (b) Boxplots of compositional-level H/C (left panel) and O/C (right panel) ratios in each Earth system. Colored dots in the boxplots are the H/C or O/C values for individual samples, and black dots indicate their mean values. Different letters (a-c) indicate a significant difference (P < 0.05) by a Kruskal-Wallis test. Regarding the organization of figure panels, we could first look at the overall view of means ± s.e of H/C and O/C ratios among all systems in panel (a) and then zoom in to the samples within each system in panel (b).

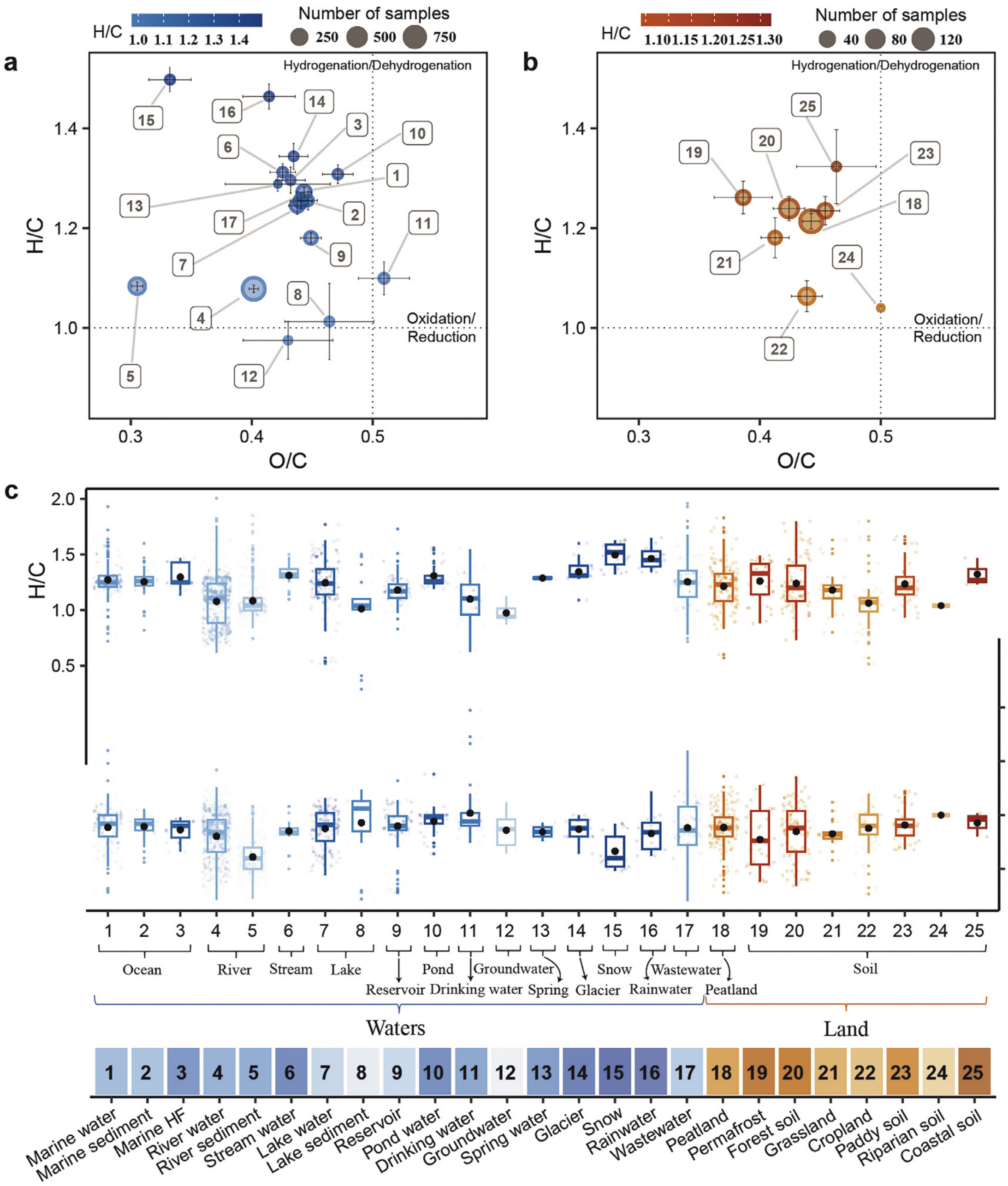

Fig. 3. Variation in H/C and O/C ratios of DOM across habitats of waters and land. Van Krevelen diagram shows the means of compositional-level H/C and O/C ratios measured by (-)ESI-FT-ICR MS in the habitats of waters (blue, a) and land (orange, b). Black dashed lines represent the direction of change in H/C and O/C ratios for chemical reactions, including hydrogenation/dehydrogenation and oxidation/reduction [12]. The number of samples in each habitat is indicated by the dot size. (c) Boxplots of compositional-level H/C (top panel) and O/C (bottom panel) ratios for better comparisons among the habitats. Significances for pairwise comparisons between habitats by a Wilcoxon test are provided in Fig. S5. Colored dots in the boxplots are the H/C or O/C values for individual samples, and black dots indicate their mean values. The dots with color gradients of blue or orange from light to dark represent H/C ratios varying from low to high, respectively. The labels with numbers 1 to 25 indicate all habitats. Marine HF: marine hydrothermal fluid. Regarding the organization of figure panels, we could first look at the overall view of means ± s.e of H/C and O/C ratios among all habitats in panels (a) and (b), and then zoom in to the samples within each habitat in panel (c).

than upland soil [47]. Different from previous studies, our synthesized global datasets, for the first time, extended such findings with unprecedently finely categorized and comprehensive habitats, which provides an overview for comparing DOM traits along the aquatic-terrestrial continuum.

3.3. Variation in DOM traits with latitude

The H/C and O/C ratios also showed predictable patterns along latitudinal gradients for the waters and land systems (Fig. 5). Specifically, H/C ratios generally showed a significant U-shaped pattern in both systems, with the lowest values occurring in latitudes of absolute 40° - 50° ($P \leq 0.05$, generalized additive models; Fig. 5a, c). This U-shaped pattern was also observed in specific habitats, such as river water, lake water, and forest soil (Fig. 5b, d). In comparison, the O/C ratios also showed a significant ($P \leq 0.05$) U-shaped pattern in the waters but a nonsignificant ($P > 0.05$) pattern in the land (Fig. 5a, c). There were also non-U-shaped patterns observed in diverse habitats, e.g., hump-shaped patterns of H/C ratios in marine water, cropland and paddy soil and a decreasing pattern of O/C ratios in paddy soil (Fig. 5). Previous studies

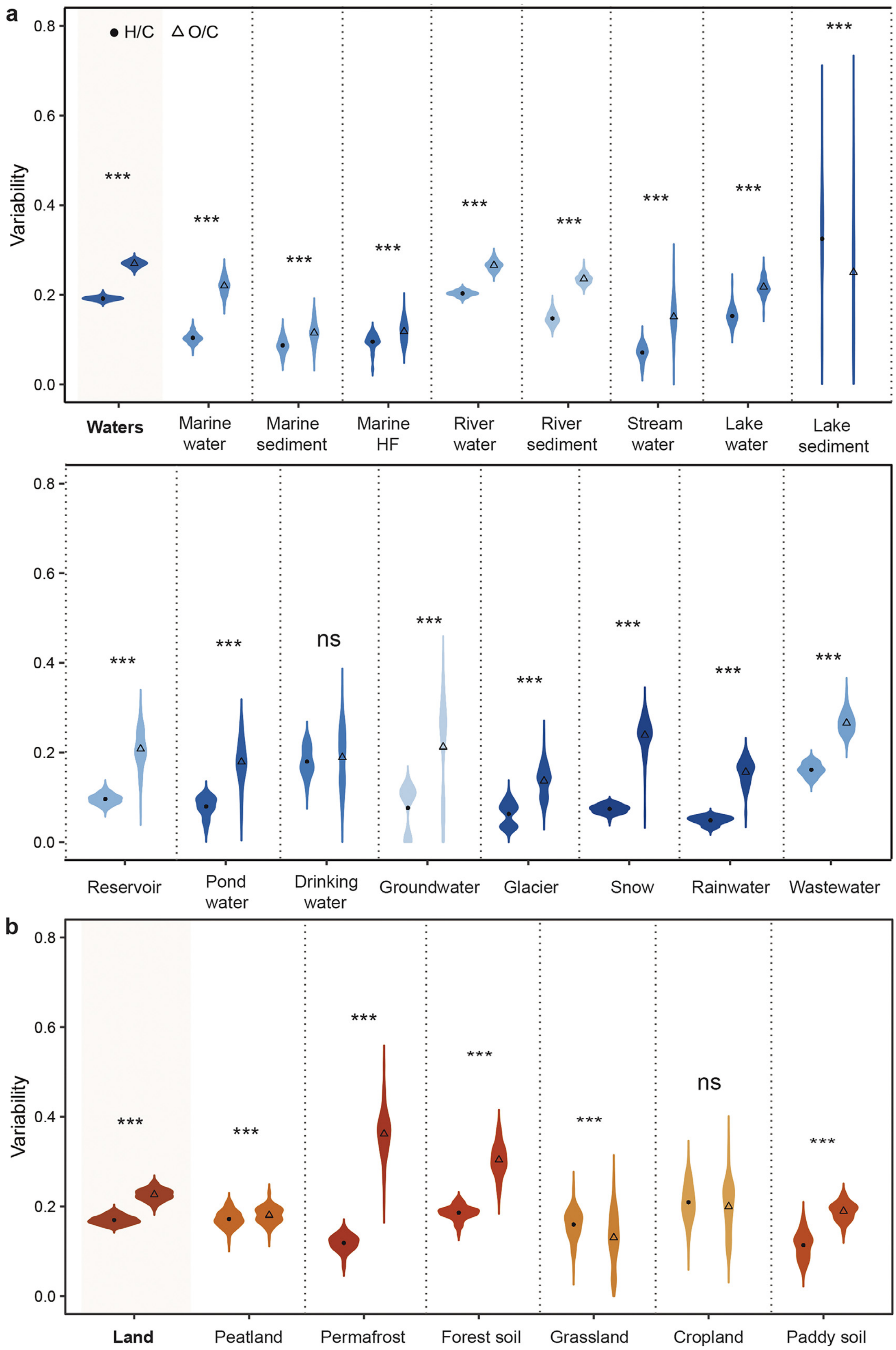

Fig. 4. Variability of H/C and O/C ratios of DOM within each habitat of waters and land. Violin plots of variability of the H/C (a) and O/C (b) ratios of DOM measured by (-)ESI-FT-ICR MS across the samples in the waters or land and their corresponding habitats. The variability was calculated as the ratio of standardized deviation and mean of the ratios by randomly selecting 50% samples (100 bootstraps) for each system or habitat. Asterisks indicate significant (***, $P \leq 0.001$) differences between two ratios by *t*-test analysis. ns: nonsignificant.

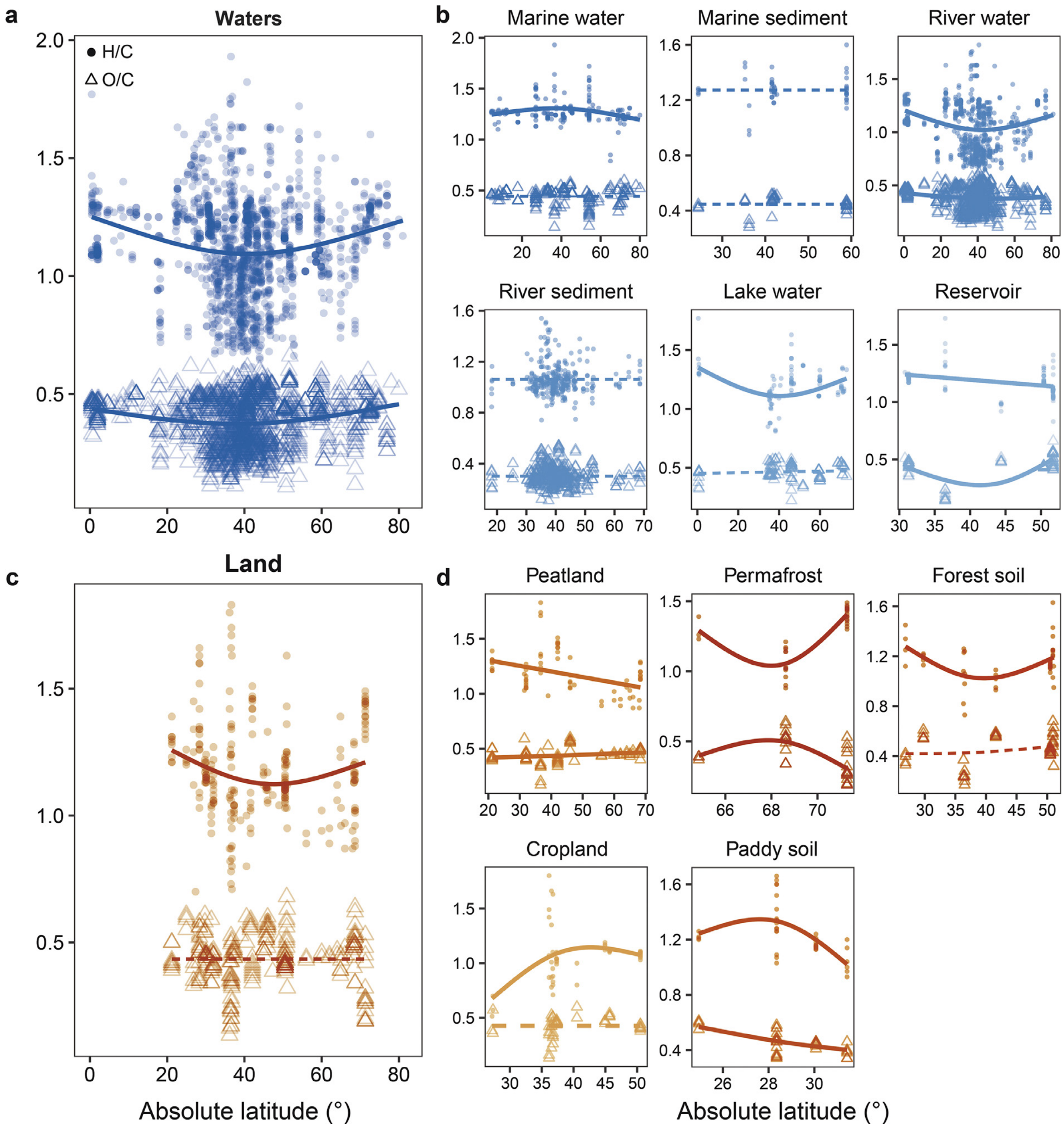

Fig. 5. The distribution patterns of H/C and O/C ratios of DOM along latitudinal gradients. We plotted the compositional-level H/C and O/C ratios measured by (-)ESI-FT-ICR MS against latitudes for the waters (blue, a) and land (orange, c) and the corresponding habitats (b, d). Latitudinal patterns are visualized with generalized additive models with 2 knots, and the significant patterns are indicated by asterisks (***, $P \leq 0.001$; **, $P \leq 0.01$; *, $P \leq 0.05$). North and South latitudes were assigned as absolute latitudes. It should be noted that it would be challenging to show the equator and polar regions in the figures, as we did not obtain samples located at latitudes of > 80°.

have shown that DOM traits have monotonically decreasing or increasing patterns along latitudinal gradients [48,49]. For example, bacterial production of fluorescent DOM in alpine and polar lakes, indicated by optical properties such as absorption at 250 nm and total fluorescence, shows a decreasing pattern within absolute latitudes of 30–75° but is lower at around mid-latitudes of ~ 50° [48]. Considering that our compiled datasets with a larger spatial scale spanned from tropics to polar regions and covered broad gradients of ecosystem properties, such as climatic and environmental factors, we show, for the first time, that H/C ratios had a predictable U-shaped latitudinal pattern for both waters and land systems. This pattern may not occur for habitats covering a small range of latitudinal gradients. Together, our results suggest that DOM is more hydrogen saturated towards the extremes at the polar regions and at the equator, while less hydrogen saturated in the mid-latitudes of 40°–50°.

3.4. Drivers of DOM traits in waters and land

The distribution patterns of H/C and O/C ratios of DOM were both significantly affected by climate and environmental variables across waters and land, as indicated by linear mixed models (Fig. 6, Figs. S6, S7, and Table S4). For the waters, environmental variables such as dissolved oxygen and ammonium had the strongest effects on the H/C ($R^2 = 0.775$, $P \leq 0.05$) and O/C ($R^2 = 0.605$, $P \leq 0.05$) ratios, respectively (Fig. 6).

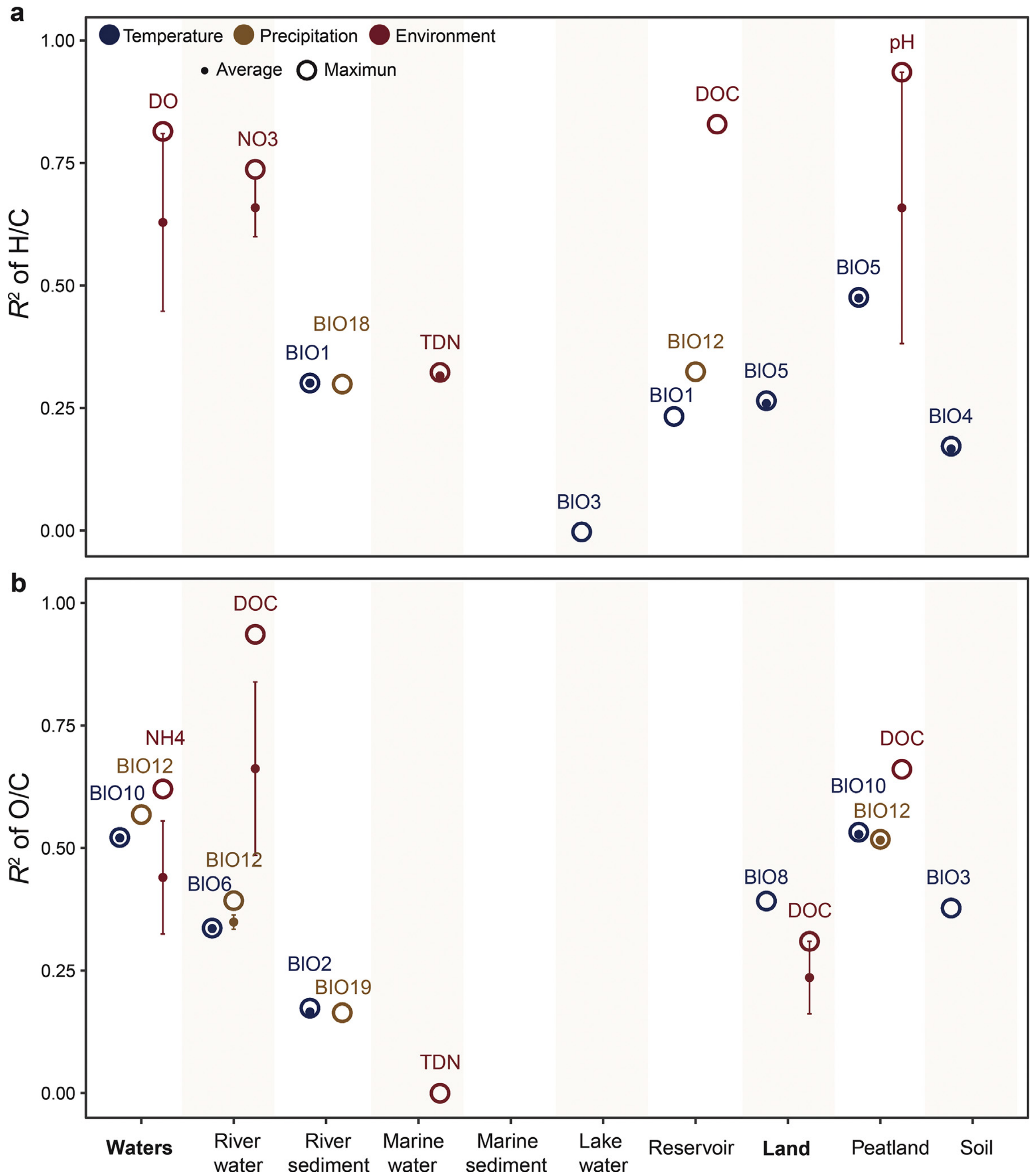

Fig. 6. The influences of climatic and environmental variables on H/C and O/C ratios of DOM. The influences of each explanatory variable on the compositional-level H/C (a) and O/C (b) ratios measured by (-)ESI-FT-ICR MS were examined with linear mixed-effects models for the waters and land and their finely categorized habitats. The significant (P < 0.05) conditionally explained heterogeneity (R^2) are shown for these variables according to their driver categories of temperature, precipitation and environments. Smaller solid dots are the average R^2 for each driver category, and open circles are the maximum R^2 among the variables in each category. We included the habitats with a sample size over 30. The abbreviations of the explanatory variables are shown in the Material and methods.

Dissolved oxygen had strong effects on H/C rather than O/C, which may be explained by the fact that high-oxygen conditions could result in the preferential degradation of high H/C compounds such as aliphatics and the production of microbial residues, while low-oxygen conditions promote the preservation of high H/C compounds such as aliphatics, sugars and amino acids, which are thermodynamically less favorable to biodegradation [22]. Climatic variables, such as mean annual precipitation and mean temperature of warmest quarter, also showed strong effects on the O/C ratios, with explained variations of 0.564 and 0.527, respectively (Fig. 6). For the land, the H/C and O/C ratios also showed significant variation explained by climatic variables, such as maximum temperature of warmest month ($R^2 = 0.332$, $P \le 0.05$) and mean temperature of wettest quarter ($R^2 = 0.425$, $P \le 0.05$), respectively (Fig. 6). Further partitioning analyses confirmed the stronger relative importance of extremes of climatic factors on H/C and O/C ratios than mean annual climates (Fig. S6). Specifically, the variances of H/C and O/C ratios were mostly explained by isothermality and precipitation of coldest quarter, respectively, in the waters, while by precipitation of coldest quarter and mean temperature of wettest quarter, respectively, in the land (Fig. S6).

However, the dominant effects of climatic and environmental factors were not always consistent across the individual habitats (Figs. 6, S7). For example, H/C and O/C ratios in marine water, reservoir water, river water, and peatland were most strongly affected by environmental variables, such as total dissolved nitrogen, dissolved organic carbon, nitrate, and pH, followed by extremes of climatic variables, such as minimum temperature of coldest month, maximum temperature of warmest month, and mean temperature of warmest quarter (Fig. 6). In contrast, the H/C and O/C ratios in river sediment and soil were dominantly affected by mean annual temperature and extremes of climatic variables such as precipitation of warmest quarter, mean diurnal range, isothermality, and temperature seasonality (Fig. 6).

These findings support previous reports showing the important roles of ecosystem properties in controlling DOM traits and decomposition rates, such as temperature [50,51], precipitation [52], carbon and nitrogen contents [53,54], and acidity [55]. Furthermore, our findings reveal additional links of DOM traits to the extremes of climatic variables beyond those drivers known from previous studies. Specifically, H/C and O/C ratios were more closely related to extremes (e.g., monthly or quarterly maximum) of temperature or precipitation than to mean annual temperature or precipitation. Earth's average temperature has increased by 1.5 °C since the preindustrial baseline, and even relatively small incremental increases in global warming (+0.5 °C) can cause statistically significant changes in extremes on the global scale and for large regions [56]. Extremes of climatic factors are key to understanding the effect of climate change on primary producers, such as plant species diversity and growth [57,58] and decomposers such as microbes [59], which would affect organic carbon characteristics. For example, maximum temperature of warmest month and mean temperature of warmest quarter had positive impacts on H/C ratios in the land system and in the habitats of peatland and soil and negative impacts on O/C ratios in waters and land systems (data not shown), likely suggesting higher decomposition caused by extreme temperature increases. Thus, our findings highlight the need to integrate extremes of climatic factors into climate change modeling when making current inferences and future predictions of organic carbon processes.

4. The implications of this study

First, our synthesized analysis provided a comprehensive survey of molecular-level perspectives of global DOM characteristics across Earth systems and climates. Trait-based metrics such as H/C and O/C ratios are relevant to the chemical reaction processes of molecules and thus effectively inform the fate of DOM, such as decomposition processes. For example, the utility of the H/C ratio as a surrogate for reactivity studies could reveal the quantifiable and comparative labile nature of DOM [18]. Our utility of these two metrics provides an overview of the current state of knowledge on the spatial distribution of DOM characteristics via a meta-synthesis approach by compiling data from the unprecedentedly finely categorized and comprehensive habitats [18,60].

Second, rather than considering these systems independent of one another, a more holistic perspective of DOM characteristics needs to be developed in a system-to-system continuum such as that in Lake Nam Co [61]. Considering that the intensity and frequency of extreme rainfall would increase with climate warming, carbon and nutrient transport from soil to inland waters and to the ocean are anticipated to increase [43,44]. Consequently, the global patterns of DOM characteristics across Earth systems or habitats could help understand how the mixing of organic carbon sources of contrasting reactivity would influence the changes in carbon cycle processes.

Third, a better understanding of molecular-level perspectives of the global DOM characteristics in response to environmental constraints is of great interest to a wide readership, as this ultimately helps understand and predict future global carbon cycle processes. It is critical to understand the global variations in DOM characteristics resulting from the spatial heterogeneity of climatic and environmental variables, which is important for estimating the potential constraints of DOM characteristics. Our synthesized analysis highlights the potential influence of climatic constraints, especially extremes of climatic factors (e.g., monthly or quarterly maximum temperature or precipitation), on DOM traits. The inclusion of these novel drivers of climate extremes could help predict DOM characteristics and further carbon cycle processes under future climate change scenarios.

5. Future perspectives

Although our mass spectrum datasets were selected considering the FT-ICR MS instrument and ESI negative ionization method, there may still be uncertainties in data comparison and interpretation across studies. For example, the differences in analytical equipment between laboratories and studies, data acquisition and processing, or sample preparation methods could be important considerations in achieving reproducible results [26]. There are also well-known limitations of unrecovered fractions during DOM extraction for FT-ICR MS, as summarized in numerous previous publications [62–64]. Our findings should be, if possible, validated using consistent measurement methods across systems and habitats. We, however, could utilize proper statistical models to account for idiosyncrasies of the data [32] and minimize the potential biases of data discrepancies across various instruments or laboratories. Furthermore, our datasets were based on the compositional-level H/C and O/C ratios of DOM. We did not consider other information in this study, such as the full dataset with individual molecules, or other trait metrics such as the number of N, P, and S, aromaticity, and nominal oxidation state of carbon. Additional trait information, especially with consistent measurement methods, would be helpful to fully understand the global carbon cycle's processing mechanisms and DOM transformations.

In addition, much larger FT-ICR MS datasets in the literature are derived from waters compared to other systems and mainly from habitats in subtropical and temperate climates. Further studies are encouraged to extend the coverage of habitats such as groundwater, spring, soils and coastal areas to a larger spatial scale spanning from tropics to polar areas. Although our main aim is to explore the important roles of climatic and environmental factors in controlling DOM characteristics across Earth systems, other potential drivers, such as microbial communities [1,10], minerals [65], and water retention time [66], have been reported to influence DOM characteristics. Future studies are encouraged to focus more on the causal relationships among these potential drivers, climate, and DOM characteristics, which will improve our understanding of the underlying mechanisms of global carbon cycle processes.

Author contributions

JW conceived the review. LH synthesized and analyzed the data with the contributions of AH and JW. AH and JW finished the first draft. JW and AH finalized the manuscript with the contributions of all authors.

Declaration of competing interest

The authors declare that they have no conflicts of interest in this work.

Data availability

The original data are available in the studies provided in Table S1. The DOM traits measured by FT-ICR MS are available on Zenodo (10.5281/zenodo.10067981). The other information is available from the corresponding author upon reasonable request.

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.fmre.2023.11.018.

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