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Key Points:

- An approach is described to extract information from convoluted spatial geochemical data
- The clustering tool applied to the Chinese marginal seas is effective in demarcating carbon impact zones that match with physical features
- Each zone has specific characteristics which are indicative of different controls on carbon dynamics

Supporting Information:

- Supporting Information S1

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Deconvolving the Fate of Carbon in Coastal Sediments

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Abstract Coastal oceans play a crucial role in the global carbon cycle, and are increasingly affected by anthropogenic forcing. Understanding carbon cycling in coastal environments is hindered by convoluted sources and myriad processes that vary over a range of spatial and temporal scales. In this study, we deconvolve the complex mosaic of organic carbon manifested in Chinese Marginal Sea (CMS) sediments using a novel numerical clustering algorithm based on ¹⁴C and total OC content. Results reveal five regions that encompass geographically distinct depositional settings. Complementary statistical analyses reveal contrasting region-dependent controls on carbon dynamics and composition. Overall, clustering is shown to be highly effective in demarcating areas of distinct organic facies by disentangling intertwined organic geochemical patterns resulting from superimposed effects of OC provenance, reworking and deposition on a shelf region exhibiting pronounced spatial heterogeneity. This information will aid in constraining region-specific budgets of carbon burial and carbon cycle processes.

Plain Language Summary In the context on ongoing climate change, it is crucial to understand how and where carbon is buried. Coastal oceans are very important areas for carbon burial globally, even though they only form a small part of the total ocean surface. These areas are very complex because there is carbon coming both from the land as well as the sea. By understanding where the carbon from land and where the carbon from the sea ends up, we can better estimate carbon storage. This paper presents a clustering approach which uses the large dataset of carbon age and concentration in the Chinese marginal seas. The clustering approach shows where the carbon from land goes and how it is buried, which areas lose carbon and which areas bury carbon. This approach could also be used in the future on other datasets such as the Arctic Seas.

1. Introduction

Coastal oceans constitute a major component in the global carbon budget (Cai, 2011; Cole et al., 2007) and form a crucial link between the terrestrial and marine carbon reservoirs (Bianchi, 2011). Increasing anthropogenic disturbances have strongly impacted the movement of carbon from land to ocean on a global scale, leading to altered terrestrial sediment fluxes (Bauer et al., 2013; Ciais et al., 2013; Keil, 2017; Regnier et al., 2013; Syvitski et al., 2003). Marginal seas are complex and dynamic areas exhibiting a high degree of spatial and temporal heterogeneity (Bao et al., 2016; Cai, 2011). Globally, delta and shelf environments represent hotspots of carbon sequestration, accounting for >85% of OC buried in modern marine sediments (Hedges & Keil, 1995). In light of ongoing anthropogenic changes in carbon stocks in coastal oceans and their possible socioeconomic ramifications (Avelar et al., 2017), it is more pressing than ever to shed light on and understand the processes governing the spatial variation in quantity and composition of OC in these shelf systems. Over the last decades, significant advances have been made in understanding biogeochemical cycles in the global oceans through classification into distinct regions based on biogeographic, geomorphic or climatological characteristics. These include approaches focused on ocean surface primary production (Longhurst et al., 1995), sinking particles intercepted by sediment traps in the water column (Honjo et al., 2008), and ocean floor sediments (Seiter et al., 2004). Related approaches have been developed for investigating sedimentary archives through geological space and time by classifying the characteristics of preserved OC into different organic facies (Jones, 1987). However, these approaches fail to capture the complexity of depositional environments that characterize continental shelf systems, and generally focus on OC content or flux as opposed to its provenance and composition. Sediment dynamics in coastal

environments are impacted by the quality and magnitude of terrestrial input as well as energy level of the shelf (Blair & Aller, 2012). In high-energy shelf environments, sediments can undergo continuous reworking where organic matter is subject to continuous oxygen exposure and extensive degradation (Aller, 1998; Aller & Blair, 2006). Despite these processes, continental shelves and margins constitute the greatest sinks of carbon in the marine realm (Hedges & Keil, 1995; Premuzic et al., 1982).

The Chinese marginal seas (CMS) constitute an important continental shelf system. Two major rivers - the Changjiang (Yangtze) and Huanghe (Yellow) - drain into the CMS, with corresponding watersheds and river networks of both fluvial systems having been strongly impacted by anthropogenic disturbances over the last millennium (Li et al., 2016; Li & Daler, 2004; Saito et al., 2001). In addition to diverse, and time-evolving inputs, complex processes operating within the CMS impart strong gradients in carbon processing and dynamics that hinder extrapolations and derivation of system-wide properties and fluxes. This complexity in the CMS clearly manifests itself in a spatially highly resolved ^{14}C dataset of surface sediment OC (Bao et al., 2016). Such high spatial resolution datasets, which are becoming increasingly attainable owing to instrumental advances which now enable facile and rapid dual carbon isotopic (^{14}C , ^{13}C) analysis coupled to elemental analyses (McIntyre et al., 2016; Wacker et al., 2013), of particular interest as they increasingly serve as a powerful tool to both constrain carbon dynamics on decadal to millennial timescales and trace of carbon sources (Drenzek et al., 2009; Griffith et al., 2010). Overall, there is a strong imperative to find effective methods that deconvolve the complex patterns in OC content and composition, and to provide a mechanistic understanding of the underlying processes responsible for this heterogeneity. In this study, we apply a clustering algorithm adapted from Nandan et al. (2017) as a novel way of classifying swaths of ocean floor into distinct "carbon impact" regions. We then apply statistical analyses to assess the region-specific factors impacting the carbon dynamics. This combination of approaches is used to deconvolve the complex carbon dynamics, yielding an improved understanding of, and quantitative constraints on carbon cycling in the Chinese coastal system. We find that this method is highly effective in elucidating the fate of terrestrial carbon in the region. Furthermore, the clustering approach provides better understanding of which areas could most benefit from further analyses, which could be a great asset for future sampling campaigns in the region.

2. Methodology

The algorithm developed by Nandan et al. (2017) for the estimation of spatially variable parameters of an Epidemic-type aftershock sequence was modified and applied to the available TOC and ^{14}C data for the CMS by assuming a log-normal distribution of these parameters (Seiter et al., 2004). Previous studies have also assumed a log-normal distribution of TOC and other parameters (Diggle & Ribeiro, 2007; Gosheva et al., 2017; Nussbaum et al., 2014; Seiter et al., 2004). Despite the unprecedentedly large ^{14}C dataset, the number of samples is insufficient to fully reject the possibility of other distribution. TOC and ^{14}C were selected as parameters because they were both the most abundant as well as most information-dense data available. Radiocarbon (^{14}C) provides information about OC material age and when combined with TOC, can provide insight into OC transport and degradation. Other data in the region was not available in the same density. The algorithm was selected because (i) the kernel could be adapted depending on the data distribution (thus accounting for the sample distribution), (ii) sampling resolution bias is minimal, and (iii) it provides contiguous clusters. In that way, it is superior to alternatives such as k-means clustering and log-Gaussian mixture models.

The essence of the method is that it uses the similarities within and dissimilarities between automatically generated regions for both TOC and ^{14}C to produce the likeliest clusters. We begin by assuming that the given spatial region (e.g. CMS in this paper) consists of q subdomains with respect to TOC and ^{14}C distribution. For this we divide the spatial region into q partitions by constructing Voronoi partitions from q randomly chosen locations (which act as Voronoi centers). Then for each partition, we estimate the mean (μ_{TOC} , $\mu_{\Delta^{14}\text{C}}$) and covariance (σ_{TOC} , $\sigma_{\Delta^{14}\text{C}}$) for a log-normal distribution of both TOC and ^{14}C using the iterative Expectation–Maximization (EM) algorithm (McLachlan & Krishnan, 2008). We repeat above-mentioned estimation of parameters for 100,000 random configurations of partitions for positive integer values of q (1–10 for this study). In order to determine the optimal set of partitions we compute a penalized log likelihood for each set of partitions as per the Bayesian Information Criterion (BIC) and choose the one with the minimum value. The BIC is a tool for model selection (Diggle & Ribeiro, 2007) and explicitly penalizes for model

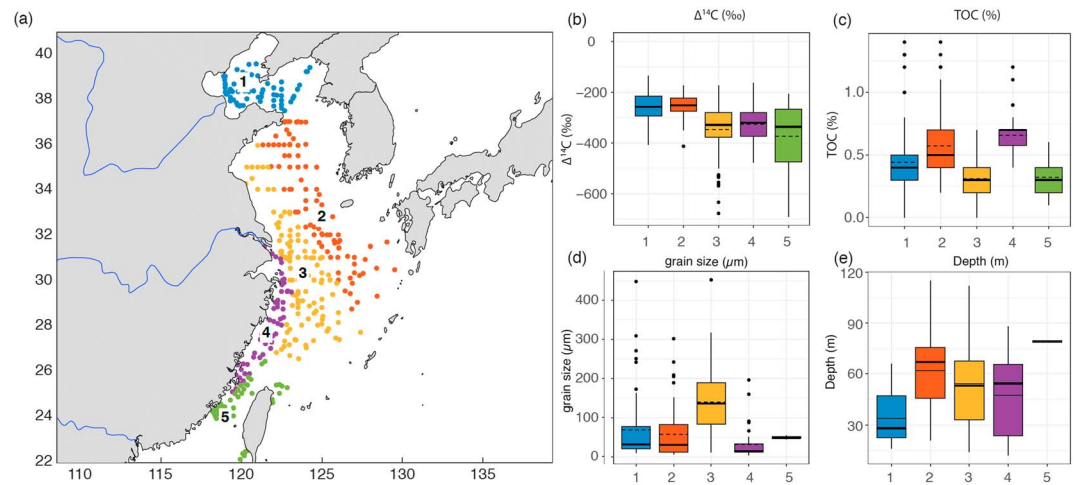


Figure 1. (a) Partitions in the CMS and (b-e) boxplots of the various measured geochemical parameters per partition as demarcated by the clustering algorithm. The regions 1–5 refer respectively to the Huanghe impact zone, the inner East China Sea-Yellow Sea (ECS-YS) shelf, the mid ECS-YS shelf, the Changjiang impact zone and the Taiwan impact zone. The bottom and top of the box present respectively the 25th and 75th percentile are indicated by the end of the colored boxes, the full band indicates the median and the dashed line indicates the average value. The whiskers indicate 1.5 times the range of between the first and third quartile for both extremes, the points are outliers. The distributions reveal geochemical differences between the partitions.

complexity (Kass & Raftery, 1995). In order to obtain a stable (robust) ensemble, set of partitions of CMS, we repeated the above-mentioned exercise 10,000 times with 357 data points chosen from the original dataset with repetition (in statistics, this process is often referred to as bootstrapping). To emphasize the commonality (similar trends) and eliminate noise in these estimated solutions, we first attributed the obtained mean and covariance values for TOC and ^{14}C to individual spatial location of the data point and then took an average of the 10,000 estimated solutions namely μ_{TOC} , $\mu_{\Delta^{14}\text{C}}$, σ_{TOC} and $\sigma_{\Delta^{14}\text{C}}$ to obtain a fuzzy (continuous) distribution of the parameters in space. For the solutions to exhibit any meaningful partitions the averaged fuzzy distribution should show a smooth but evident spatial variation of parameters with each distinct spatial zones. These zones can be observed as zones of similar μ_{TOC} , $\mu_{\Delta^{14}\text{C}}$, σ_{TOC} and $\sigma_{\Delta^{14}\text{C}}$ values. Based on this spatial variation of parameters we proceed to manually identify distinct spatial regions in CMS and compute their corresponding BIC. We observe that the BIC thus obtained shows a lower value than the best solution obtained through a random search of partitions (100,000), and thus we recognize it as the optimal partition set.

For this optimal partition set, Spearman correlation (significant for $p < 0.05$) was performed on the data for which all variables were available (n_{complete} , i.e. with TOC, ^{14}C , depth and grain size data) using the statistical software R (version 1.0.44 © 2009–2016 RStudio, Inc). For the dataset explored here, there is less depth and grain size data available as compared to ^{14}C and TOC data. Further details of the method can be found in the supplementary information (SI) 1–4 and supporting information Figures S1–S4. Regarding code availability, the analyses in MATLAB are available upon request.

3. Results and Discussion

3.1. Region-specific Carbon Dynamics

The numerical clustering based on ^{14}C and TOC data resulted in a sub-division of the CMS into five regions, each with distinct geochemical characteristics (Figure 1). These five regions are defined as the Huanghe impact zone, the inner East China Sea-Yellow Sea (ECS-YS) shelf, the mid ECS-YS shelf, the Changjiang impact zone and the Taiwan impact zone.

The first region (Huanghe impact zone; $n_{\text{complete}} = 60$) encompasses the outflow of the Huanghe (Yellow River) to the shallow Bohai sea (Deng et al., 2006). It is characterized by relatively high $\Delta^{14}\text{C}$ values (mean -257 ‰) at shallow depths (mean 34 m) (Figure 2). The ^{14}C -depleted or relatively old carbon

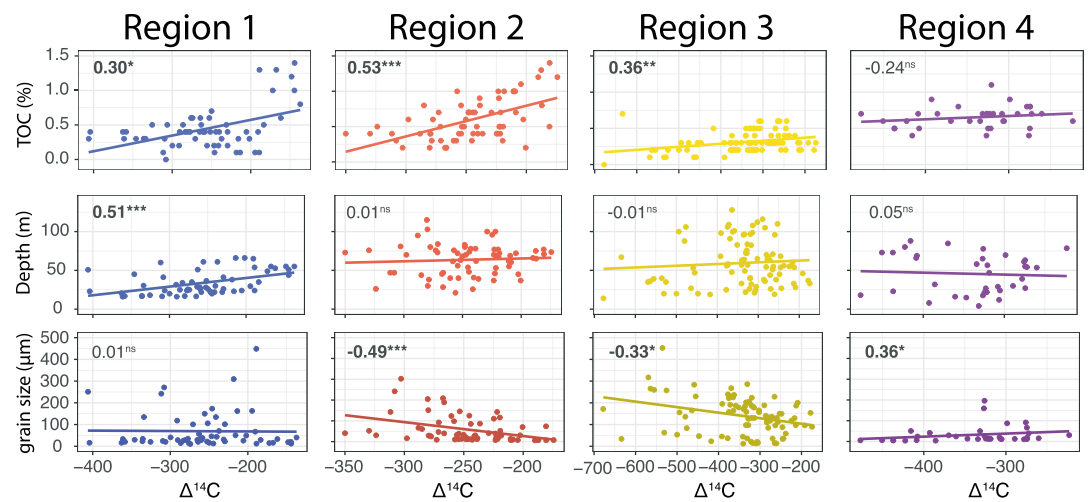


Figure 2. Relationship of grain size (μm), depth and TOC with $\Delta^{14}\text{C}$ for regions 1–4. Lines represent linear regression, indications of p indicate significance ($p < 0.05$, *) ($p < 0.005$, **) ($p < 0.0005$, ***) or not significant (ns) results following Spearman correlation.

signature may reflect inputs of pre-aged carbon from the Huanghe which entrains material from the extensive loess deposits on the Tibetan plateau (Tao et al., 2015). $\Delta^{14}\text{C}$ correlates positively with TOC (0.30*) and depth (0.51***). The positive correlation of $\Delta^{14}\text{C}$ with TOC and depth could either be due to enhanced reworking and resuspension (Hedges et al., 1999) of sediment at shallower depths owing to tidal action, or to larger contributions from ^{14}C -enriched organic matter marine primary production (Tang et al., 2003) at greater depths.

The second region (Mid ECS-YS shelf; $n_{\text{complete}} = 66$) and is marked by the relatively high $\Delta^{14}\text{C}$ (mean -252 ‰), TOC (mean 0.6%) and the greatest average water depth (68 m). $\Delta^{14}\text{C}$ values also positively correlate with TOC (0.53**), and negatively with grain size (-0.49 ***) (Figure 2). These relatively high TOC and $\Delta^{14}\text{C}$ values are consistent with a regional depocenter for fine-grained sediments (Zhu & Chang, 2000). Accumulating sediments may entrain relatively young organic carbon from marine productivity supported by supply of nutrient-rich upwelling waters from the Kuroshio current (Chen, 1995; Wu et al., 2003), as well as terrestrial materials into this region from the eroding inner margin (Zhu & Chang, 2000). The negative correlation of $\Delta^{14}\text{C}$ with grain size may reflect diminished organic matter protection on mineral surfaces (Aller, 1998), or sediment winnowing processes (Hedges et al., 1999), each resulting in older and lower TOC abundances on larger particles. Blair and Aller (2012) explain that low-energy coastal environments of particular significance in the global carbon cycle, being responsible for approximately 40–50% of OC accumulation in coastal zones. In this context, it is likely that the Mid ECS-YS shelf region represents a key locus for the storage of carbon in the CMS.

The third region (Inner ECS-YS shelf; $n_{\text{complete}} = 88$) is characterized by more ^{14}C -depleted (older) carbon (mean -346 ‰), low TOC contents (mean 0.3%), and a large spread in grain size (mean 140 μm), and thus rendering it geochemically distinct from the mid shelf region. This region approximately demarcates a previously mapped sandy swath of the East China Sea shelf (Niino & Emery, 1961). Although the inner shelf region exhibits a positive correlation of $\Delta^{14}\text{C}$ with TOC (0.36*) and negative correlation with grain size (-0.33 *), these correlations are markedly weaker than in the other regions (Figure 2). The weaker correlations, low TOC content and large range of $\Delta^{14}\text{C}$ values could potentially be due to high(er) energy-levels of this shelf area, which would lead to increased resuspension of sediment, prompting increased OET that, in turn, leads to lower TOC content. Coastal oceans can serve as sedimentary organic matter “incinerators” (Aller & Blair, 2006; Yao et al., 2014), where material is extensively and intensively reworked and respired. The large grain sizes present may indicate that there is a significant marine input, potentially from foraminifera settling down from production in overlying surface waters (Gong et al., 2003). Alternatively, coarser grains could be terrestrially-derived, and concentrated through hydrodynamic sorting processes (Bianchi et al., 2007).

The fourth region (Changjiang impact zone; $n_{complete} = 36$) encompasses the area proximal to the Chinese mainland and enveloping the Changjiang outflow and sedimentary plume direction shaped by the Zhejiang Fujian Coastal Current (ZFCC). This includes an extensive mobile mud belt that runs parallel to the Chinese coastline (Liu et al., 2007; Niino & Emery, 1961) which is marked by relatively ^{14}C -depleted organic carbon (mean -326‰), fine-grained sediments (mean $32\ \mu\text{m}$) and elevated TOC contents (mean 0.7%). This signature could be a consequence of pre-aged organic matter emanating from the river (Li et al., 2014; Wang et al., 2012), and additional aging due to sediment incineration (Aller & Blair, 2006). In the Changjiang impact zone, $\Delta^{14}\text{C}$ only exhibits a significant correlation (0.36^*) with grain size (Figure 2) and, in contrast to the other impact zones, there is no correlation between $\Delta^{14}\text{C}$ and TOC. This may imply that the recorded signal is primarily of terrestrial origin, an interpretation supported by the fine-grained nature of the sediments relative to other regions of the CMS (Figure 1e). The positive correlation of $\Delta^{14}\text{C}$ and grain size implies that smaller particles tend to be older, which could potentially be due to a terrestrially-influenced signal, e.g. from fine-grained sedimentary rock (e.g., petrogenic) debris (Wu et al., 2007). In addition to rock-derived inputs, other pre-aged biospheric material (Tao et al., 2015) as well as young material (both biospheric and marine) could also contribute to plume, however further studies (e.g. ^{13}C , biomarkers) are required to derive definitive conclusions.

The fifth region (Taiwan impact zone; $n_{complete} = 2$), which partially overlaps with the Taiwan Warm Current (TWC) and is located between the Chinese mainland and Taiwan island, is marked by the oldest carbon (mean $\Delta^{14}\text{C} -373\text{‰}$). Unfortunately, the lack of data points in this region precludes rigorous statistical analysis and assessment of correlations. Moreover, sharp differences within this dataset preclude quantitative analyses, likely owing to the distinctive characteristics of OC associated with sediments rich in petrogenic OC supplied from Taiwan (Bao et al., 2016; Kao et al., 2014). The impact of these inputs are superimposed on those due to hydrodynamic sorting, Changjiang inflow, marine productivity, conspiring to limit the success of clustering analysis in this region. Further investigations, both with regard to increasing sampling density and areal coverage would be necessary in order to effectively demarcate and identify drivers associated with this impact zone.

Further characterization of these impact zones could be gained from procuring additional geochemical and sedimentological information. For example, parameters such as $\delta^{13}\text{C}$ could provide information about input to the sediment from primary oceanic production (Wu et al., 2003), while surface area and C:N ratios could shed light on the degree of degradation (Blair & Aller, 2012). Carbonate content is a traditional indicator of marine input from biocalcifying organisms (Niino & Emery, 1961), while biomarker information could further constrain organic matter provenance and age (Eglinton & Eglinton, 2008; Tao et al., 2016).

3.2. Carbon Impact Zones

Classifications of different ocean regimes according to ecosystem properties have proven a useful framework for assessing underlying biogeochemical processes (Honjo et al., 2008; Longhurst et al., 1995; Seiter et al., 2004). However, coastal oceans present particular challenges due to their intrinsic complexity, with diverse processes operating over different spatial and temporal scales. Implementation of Regional Oceanic Modeling System (ROMS) approaches can simulate the dynamic coastal environment (Gruber et al., 2012), however, these types of models are primarily focused on water column processes and presently do not capture the intricacies of sedimentation at the seafloor. In contrast, sediment-centric investigations (e.g. Jones (1987)) define depositional facies according to general sedimentological and stratigraphic principles. Seiter et al. (2004) adopted a variation of this approach, classifying sedimentary provinces in the global ocean based on the TOC content of sediment. While the organic facies concept has the benefit that it can be applied across geological time, thereby linking actively forming and ancient sedimentary settings, TOC content remains a rudimentary criterion for defining specific depositional regimes given the myriad inputs and pre- and post-depositional processes that characterize shallow continental margin environments. The self-clustering approach adopted here harnesses information held not only in TOC content (Seiter et al., 2004) but also the ^{14}C signature of the TOC in order to resolve sources and processes contributing to the spatially heterogeneous mosaic of carbon signals that characterize marginal sea sedimentary environments. The latter provides key constraints on carbon sources and the effect of sediment reworking on organic matter composition (Bao et al., 2016; Tao et al., 2015). Combining both geochemical characteristics, we delineate the CMS system into distinct “carbon impact zones” that take into account both carbon provenance and sedimentation

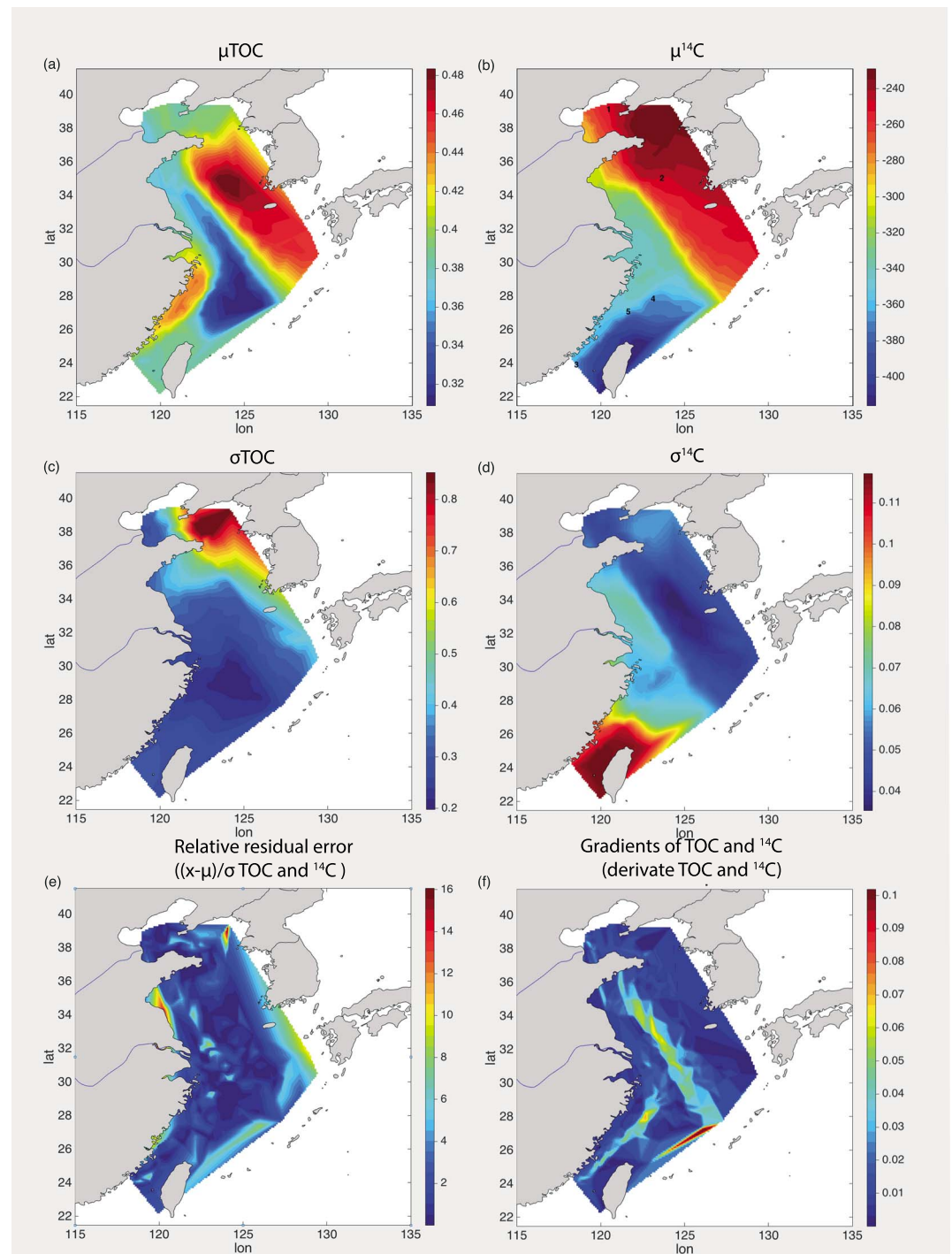


Figure 3. average modelled values (μ) for (a) TOC and (b) ^{14}C and the variance (σ) for (c) TOC and (d) ^{14}C , (e) the relative residual error as indicated by the difference of the measured value and the calculated mean normalized to the variance of TOC and ^{14}C $[(x-\mu)/\sigma]$ and (f) areas where largest gradients occur, as indicated by the added derivative of the average (μ) TOC and ^{14}C values.

processes and afford a greater degree of spatial resolution than hitherto available (Honjo et al., 2008; Longhurst & Glen Harrison, 1989; Seiter et al., 2004), providing a powerful new framework for constraining regional carbon budgets. Other studies have examined spatial patterns of organic carbon and movement of sediment in the Chinese coastal ocean (Li et al., 2016; Liu et al., 2006, 2007; Saito et al., 2001; Zhu &

Chang, 2000). Although most studies focus on specific regions, there is concordance with the carbon impact zones found in this paper. River-dominated areas, such as the Huanghe (region 1) and Changjiang (region 4) impact zones, echo the movement and deposition of sediments supplied by the rivers as described by Saito et al. (2001), Liu et al. (2006, 2007) and Niino and Emery (1961). Furthermore, the fine-grained muds of the shelf as described by Li et al. (2016) and Niino and Emery (1961) match well with the Mid ECS-YS shelf (region 2). Additionally, the Inner ECS-YS shelf matches the areas where higher sediment erosion rates have been observed (Zhu & Chang, 2000). Lastly, the Taiwan-proximal zone is marked by high terrestrial carbon input and burial with particularly high inputs of petrogenic carbon (Kao et al., 2014). We believe the delineation of these carbon impact zones is particularly robust because it is based on bootstrapping (10,000 times) of the data (more details in supporting information and supporting information Figure S4).

3.3. Optimized Sampling

Spatial modeling can be utilized to identify specific areas that would benefit most from future sampling and analysis (Diggle & Ribeiro, 2007). Specifically, areas that require higher sampling density include regions where (i) the variance (i.e., standard deviation) is high, (ii) there is a large residual error (i.e., large discrepancy between modeled and measured value) and (3) spatial gradients are steep (i.e., where values of TOC or ^{14}C change rapidly). In this study, the clustering approach estimates the mean (μ) and variance (σ) for both $\Delta^{14}\text{C}$ and TOC for all points in the ECS (Figure 3a-d), with average values of TOC demarcating two elevated-carbon depocenters - on the mid shelf and south of the Changjiang. The $\Delta^{14}\text{C}$ signature shows an abrupt divide between relatively young material in Huanghe region and the mid shelf (regions 1 and 2) and older material in regions from the inner shelf southwards (regions 3–5). The variance of TOC is high in the northernmost Yellow Sea, while variance of ^{14}C is highest towards the south, near Taiwan. As an illustration of information embedded in this uncertainty, we note the residual error normalized to the variance (Figure 3e) shows higher uncertainty around the sparsely populated edges of the ECS, as well as in the vicinity of Taiwan and at the transition between the inner and mid ECS-YS shelf. The gradient map (derivative of μ estimates of both TOC and ^{14}C) also reveals that the signature changes rapidly between the inner and mid ECS-YS shelf (regions 2 and 3, respectively) and around the Changjiang plume (region 4) (Figure 3f). Regions that stand out in these respects are (i) around Taiwan and (ii) the outer ECS regions, the (iii) transitions between the inner and middle shelf and (iv) the area around the Changjiang-derived mud belt (Liu et al., 2006).

4. Conclusion

The self-clustering approach described here based on ^{14}C and TOC data serves as an effective tool to identify various carbon impact zones, each defined by distinct characteristics that reflect a combination of OC inputs and processes (e.g., currents) in the CMS. This provides a novel means to classify ocean regions that is both complementary, and provides a bridge, to those focusing on surface ocean and water column properties. In the present case, we harness diagnostic information embedded in the concentration and radiocarbon content of OC residing in surface sediments. We show that the controls on sedimentation signature are highly spatially variable, implying that development of accurate carbon budgets and interpretation of sedimentary records for such marginal sea systems requires further sub-division into discrete zones. Utilization of the two criteria adopted here (TOC and ^{14}C) results in the deconvolution of the complex mosaic of organic matter properties observed by Bao et al. (2016) into five distinct “impact zones”. These zones each have geochemical characteristics with different factors contributing to the distinctive geochemical signatures including carbon and sediment sources, as well as reworking and redistribution processes. There is a strong contrast between the inner and mid ECS-YS shelf, which respectively constitute areas of (terrestrial) carbon incineration and deposition. There are areas which demarcate direct riverine input and influence of coastal water movement, such as the tidal reworking on the Huanghe-derived sediments and the reworking of Changjiang-derived sediment along the ZFCC current which forms the mud belt. The impact of Taiwan as a point source of terrestrial sediment is also apparent. While this novel clustering approach has thus far only been applied to the CMS system, it is both scalable and applicable to other regions as further high-spatial resolution datasets become available. Furthermore, spatial analysis of residual errors serves to highlight those zones that would benefit most from more intensive sampling and investigation. Similar clustering

approaches could potentially also be applied to deeper-time sedimentary archives (Eglinton & Eglinton, 2008) and help elucidate trends and thus aid the investigation of past changes of environmental conditions.

Author contributions

Tessa van der Voort structured the project and research questions, Utsav Mannu led the numerical clustering, associated preparations and aided in the method section. Thomas Blattmann helped with the interpretation of the results. Rui Bao and Meixun Zhao provided region-specific input. Timothy Eglinton guided in the paper development and paper construction. Tessa van der Voort prepared the manuscript with the help of all co-authors.

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