The impact of pre-restoration land-use and disturbance on sediment structure, hydrology and the sediment geochemical environment in restored saltmarshes.

Kate L Spencer*, Simon J Carr, Lucy M Diggens, James A Tempest[1], Michelle A Morris, Gemma L Harvey

*Corresponding author email k.spencer@qmul.ac.uk

School of Geography, Queen Mary University of London, Mile End Road, London E1 4NS, UK.

[1]Present address: Department of Geography, University of Cambridge, Downing Place, Cambridge, CB2 3EN

Abstract

Saltmarshes are being lost or degraded as a result of human activity resulting in loss of critical ecosystem services including the provision of wild species diversity, water quality regulation and flood regulation. To compensate, saltmarshes are being restored or re-created, usually driven by legislative requirements for increased habitat diversity, flood regulation and sustainable coastal defense. Yet, there is increasing evidence that restoration may not deliver anticipated ecosystem services; this is frequently attributed to poor drainage and sediment anoxia. However, physical sediment characteristics, hydrology and the sediment geochemical environment are rarely examined in restoration schemes, despite such factors being critical for plant succession.

This study presents the novel integration of 3D-computed X-ray microtomography to quantify sediment structure and porosity, with water level and geochemical data to understand the impact of pre-restoration land use and disturbance on the structure and functioning of restored saltmarshes. The study combines a broad-scale investigation of physical sediment characteristics in nine de-embanked saltmarshes across SE England, with an intensive study at one site examining water levels, sediment structure and the sediment geochemical environment.

De-embankment does not restore the hydrological regime, or the physical/chemical framework in the saltmarshes and evidence of disturbance includes a reduction in microporosity, pore connectivity and water storage capacity, a lack of connectivity between the sub-surface environment and overlying floodwaters, and impeded sub-surface water flow and drainage. This has significant consequences for the sediment geochemical environment. This disturbance is evident for at least two decades following restoration and is likely to be irreversible. It has important implications for plant establishment in particular, ecosystem services including flood regulation, nutrient cycling and wild species diversity and for future restoration design.

Keywords: porosity; managed realignment; de-embankment; microtomography; drainage; ecosystem service

1. Introduction

Saltmarshes are globally important environments occupying c. 5.1 Mha of the Earth's surface (Pendleton *et al.*, 2012) and providing a range of ecosystem services (Costanza *et al.*, 1997; Millennium Ecosystem Assessment, 2005). However, saltmarshes are threatened by sea level rise, human population growth, urbanization and pollution, causing degradation or loss of habitat worldwide. This can result in the loss of critical ecosystem services including the provision of nursery habitats, coastal defense and detoxification (Barbier *et al*., 2011). To compensate, a large number of coastal restoration projects have been implemented in recent decades, frequently driven by legislative requirements for improved biodiversity e.g. the EU Habitats Directive and Birds Directive (European Parliament and the Council of the European Union, 1992), and for sustainable coastal defense and flood storage (Esteves 2013). Increasing evidence suggests that restored saltmarshes, recreated through reversion to tidal inundation of previously drained and defended land, have lower biodiversity and ecosystem service delivery than anticipated (e.g. Mazik *et al.,* 2010; Mossman *et al.*, 2012; Esteves 2013; Brooks *et al*., 2015) and that, whilst environmental enhancement has been achieved, there may be consequences for ecosystem functioning (Doherty *et al.*, 2011). Therefore, in order to improve the delivery of ecosystem services, there is a critical need to understand both the structure and function of restored saltmarshes.

Restoration aims to recover saltmarsh ecosystem structure and function to reference conditions and assumes that as long as the physical and/or-chemical structure of the system is restored, colonization by saltmarsh plants should follow (Borja *et al.*, 2010). Surface elevation of saltmarshes is considered the most important physical/structural parameter in restoration design (Howe *et al.*, 2009), having a direct influence over plant colonization through controlling the hydroperiod and hence sediment aeration. The hydroperiod concept, defined as the proportion of time for which a wetland is submerged (Mitch and Gosselink 2007), is however over-simplistic with water depth, tidal regime, frequency of tidal flooding, distance to creek drainage networks and precipitation/evapotranspiration all influencing sub-surface saturation and net water flux (Ursino *et al.*, 2004; Eaton and Yi, 2009; Spencer and Harvey 2012; Xin *et al.*, 2013a, 2013b). The flux of pore water through the sub-surface environment is also critical for controlling abiotic conditions in the sediment including redox status, nutrient availability, salinity and the presence of potentially toxic S^-, Mn^{2+} and Fe^{2+}, all of which may be as critical as the hydroperiod for determining plant growth and ecological zonation (Silvestri et al. 2005; Wolters *et al.*, 2008; Smith *et al.*, 2009; Erfanzadeh *et al*., 2010; Howe *et al.*, 2010; Engels *et al.*, 2011; Davy *et al.*, 2011; Xin *et al.*, 2013a, 2013b; Wilson *et al.*, 2015). Flux and transport pathways of pore water through the sub-surface environment and surface-groundwater interactions are also controlled by the sediment texture (e.g. porosity) and structure (e.g. stratigraphy) (Xin *et al.*, 2009; Wilson and Morris, 2012). Yet, whilst they may potentially be important physical system parameters that influence ecosystem structure and function in restored saltmarshes they are rarely studied.

Saltmarshes are frequently re-created from land that has previously been embanked to prevent coastal inundation and drained, usually for agricultural purposes. This results in a legacy of significant pre-restoration disturbance including de-watering, compaction and mineralization of organic matter. Such disturbance also impacts sediment structure, including the collapse of pore space (Hazelden and Boorman, 2001; Boorman *et al.*, 2002; Ellis and Atherton 2003) resulting in poor drainage (Crooks *et al.*, 2002; Grismer *et al.*, 2004; Montalto *et al.*, 2007; Tempest *et al.*, 2015) and this pre-restoration disturbance may trigger saltmarsh recovery towards an alternative state, as the speed and rate of ecosystem structure recovery may be dependent upon such abiotic factors (Moreno-Mateos *et al.*, 2012). Therefore, even though it is recognized that poor drainage and sediment anoxia may be responsible for poor species composition in restored saltmarshes (Mossman *et al.*, 2012) there is a general lack of understanding of the impact of restoration on sedimentary processes (Esteves 2013). Furthermore, there has been little detailed investigation of sediment structure and how this may influence sub-surface hydrology and the sediment geochemical environment.

This study investigates the impact of pre-restoration land use and disturbance on sediment structure in saltmarshes restored through de-embankment, and explores the implications for sub-surface hydrology and the sediment geochemical environment as limiting factors for the successful restoration of saltmarshes. A novel combination of 3D structural sedimentology, geochemistry and hydrological datasets permits evaluation of the sediment structural controls over saltmarsh restoration at a number of locations in southeast England and considers whether structural characteristics, including the hydrological regime, of the saltmarsh have been restored.

2. Methods

A combination of two spatial scales of investigation were adopted for this study. A broad-scale approach analyzed physical sediment characteristics across a number of restoration sites in southeast England (Figure 1, Table S1), representing saltmarshes that were either naturally de-embanked due to storm surges in the c. 1890s and the mid-20th Century or deliberately through removal of a hard sea defense to allow tidal inundation (managed realignment - MR). At each site matched-pair data were collected from adjacent undisturbed saltmarshes, where sites were matched in terms of elevation (+/- 10 cm) and hence hydroperiod, and distance from creek edges. These will be referred to as ‘de-embanked’ (DE) and ‘natural’ (N) saltmarshes respectively. A more intensive study, including detailed sediment structural analysis, as well as pore water and sediment geochemistry, and sub-surface hydrology was also carried out at Orplands Farm MR site (Figure 1), an 11-hectare site de-embanked in 1995 to allow the tidal inundation of former agricultural land.

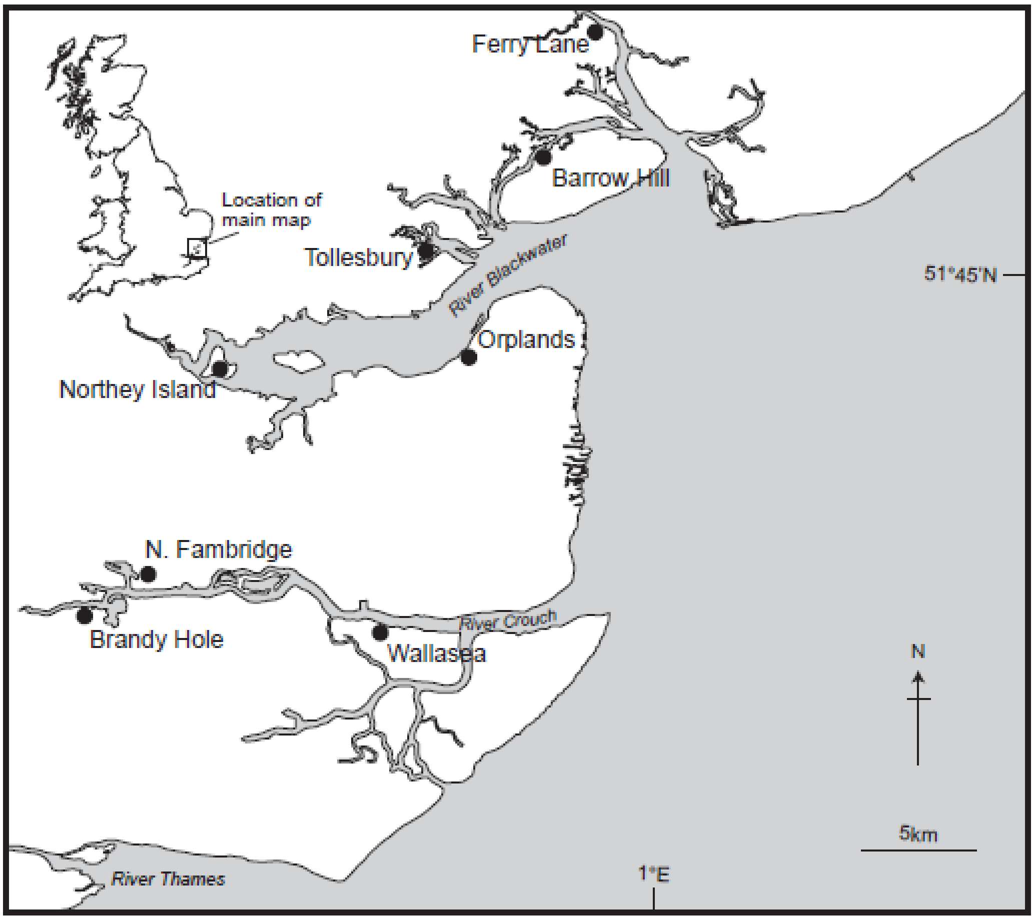
Table S1: Site locations for matched pairs of natural and de-embanked saltmarsh

Site location	Method of de-embankment	Breach date
Northey	Historical storm surge	1897
North Fambridge		1897
Brandy Hole		1897
Barrow Hill		1953
Wallasea		1953
Ferry Lane		1945
Tollesbury	Managed Realignment	1995
Orplands Farm		1995
Northey		1991

2.1. Broad-scale Investigations – physical sediment characteristics

To examine the broad-scale physical sediment characteristics of saltmarshes, sediment cores were collected at eight sites in Essex, southeast England (Figure 1). At one site (Northey) the saltmarsh had been de-embanked both historically during storm surges and via MR, resulting in two matched pairs at this location, and nine matched pairs in total for the study (Table S1). At each site, three sampling locations were selected randomly along a transect at the same elevation (+/- 10 cm) and six 30 cm sediment cores were collected using a 3 cm diameter gouge corer (18 replicates in total). Sample cores were extracted, wrapped in film and transported back to the laboratory and refrigerated at 4 °C until required. Nine of the cores remained intact and measurements of sediment characteristics were made on the bulk 30 cm core. The other nine cores were sub-divided into 5 cm increments to measure sediment characteristics with depth. Moisture content (MC) (measured following drying at 105°C overnight and calculated relevant to sediment dry weight), % loss on ignition (LOI) (measured following combustion at 550°C) and dry bulk density (measured by calculating the total sample volume and mass following drying at 105°C) were calculated for each sample.

Figure 1: Location of sampling sites in south east England.



2.2. Intensive-scale investigations at Orplands Farm MR site

Additional sediment cores from both the de-embanked and natural saltmarsh at Orplands Farm were recovered at the same locations as sampled for the broad-scale study (Figure 1). Sampling and analysis of physical sediment structure, and sub-surface water levels was carried out in 2012 and 2013, whilst pore water geochemical data were collected in 2010. In addition, sediment geochemical data from an earlier study (2005) were also re-examined. All sampling was undertaken at low or falling tide conditions during winter months.

Physical Sediment Structure and Porosity

Deeper (up to 1.4 m) cores were collected and described in the field. In addition, triplicate sediment cores were recovered using the advanced trimming method of Hvorslev (1949) by inserting clear plastic tubes (44 mm internal diameter) into the saltmarsh surface to a depth of 15 cm. This approach minimized potential disturbance to sediment structure during recovery. Sealed sediment cores were described in the field and transferred to the laboratory within 4 hours and stored at 4 °C until required. During transport and storage the cores were held in a vertical position to minimize potential disturbance.

Each sediment core was examined within 2 days of sample collection using X-ray microtomography (μCT), a non-destructive technique capable of producing three-dimensional models derived from the attenuation of X-ray energy by samples, dependent upon a combination of material bulk density and atomic number with higher attenuation representing high-density and/or high atomic number materials (Ketcham and Carlson, 2001; Cnudde and Boone, 2013). Scans were undertaken on all sealed core tubes on sediment collected between 20 - 100 mm deep to exclude the irregular saltmarsh surface and to explore the immediate sub-surface sediment characteristics in each sample. Earlier estimates of sediment accumulation rates at this site (Spencer *et al.*, 2008) indicate that the pre-breach land-surface should be at c. 4 cm depth, overlain by inter-tidal sediment deposited post-breach, and hence both pre- and post-restoration sediment facies should have been imaged using this approach. Samples were scanned using a Nikon Metrology XT H 225 X-ray CT system, with a Perkin Elmer XRD 0820 CN3 16-bit flat panel detector (Nikon Metrology, Tring, Hertfordshire, UK). Scans were performed using Inspect-X (Quiggin, 2011) for X-radiogram acquisition, with reconstruction completed using CTPro (Ray, 2011) resulting in cubic volumetric 3D models of 1024 x 1024 x 1024 voxel dimensions with a voxel size of 76.0 μm. Scanning and reconstruction settings were consistent for all samples. Visualization of reconstructed 3D models was undertaken using Drishti 2.1 volume rendering software (Limaye, 2006; 2012), which is highly suited for visualizing heterogeneous samples which are often challenging to segment into their key components (e.g. Bendle *et al.*, 2015). Drishti was used to visually identify and segment the bulk-phases of each sediment core on the basis of X-ray energy attenuation allowing interpretations of sediment components (e.g. pore space, mineral particles, sediment matrix and organic matter) to be drawn. Each 3D volume was further sub-sampled into 6 equal size depth increments (labelled A - F in each volume) for detailed analysis of porosity (Table S2) using BoneJ and FIJI open-source image analysis software (Doube *et al.*, 2010; Schindelin et al., 2012).

Table S2: Parameters assessed in intensive scale studies.

Property	Key Characteristics	Reference
% Porosity	Measure of bulk sediment porosity; measured through volume fraction tool (BoneJ)	Doube *et al*. (2010)
Euler-Poincaré Characteristic	Measure of network connectivity and redundant pore connections: 'Directness' of pore system	Vogel (1997)
Pore Network Anisotropy	Measure of the consistency of directional organisation of pore network	Odgaard (1997)
Mean Branches per Pore	Measure of pore network complexity	Polder *et al*. (2010)

Hydrology - sub-surface water levels

To explore the impact of disturbance and sediment structure on sub-surface water levels, piezometers and pressure transducers were installed in triplicate on both the natural and de-embanked site at Orplands Farm. Creek networks can effect sub-surface saturation and lateral sub-surface flows (e.g. Xin *et al.*, 2013a; 2013b), and therefore the piezometers were installed > 15 m from the edge of the nearest creek edge in both the natural and de-embanked sites to minimize impact (Hemond and Fifield 1982). Piezometer wells were dug to a depth of 60 cm and a piezometer (200 cm x 4.4 cm) was then placed into each well. All six piezometers were constructed out of ABS (plastic) with the screen located along the bottom 40 cm of the piezometer. The entire length of the screen was perforated (ø 0.5 cm) at 10 cm vertical intervals. Pressure transducers (Solinst 3001, level-logger) were hung using laminated wire and positioned 5 cm from the bottom within all six piezometers. To ensure the piezometers were not directly flooded the piezometer wells were capped and sealed with bentonite. A barologger was also installed above ground taking measurements of air-pressure. All pressure transducers took simultaneous measurements at 15-minute intervals over a period of 2 months in 2012. In order to compare water-levels between the natural and de-embanked saltmarshes, surface elevation relative to ordnance datum (O.D.) of all six piezometers was obtained using a d-GPS (Topcon, +/- 3 mm).

Sediment and pore water geochemistry

In order to examine the impact of disturbance, through drainage, agriculture and de-embankment, on the sediment geochemical environment both sediment and pore water chemistry was examined. Sediment geochemistry was examined in two sediment cores collected from the natural and de-embanked saltmarsh (40 and 20 cm depth respectively) using PVC tubing, returned to the laboratory and sub-divided into 2 cm increments before being dried, crushed and digested using a microwave assisted (CEM MARSX 2455 MHz) Aqua Regia extraction (Bettinelli *et al.*, 2000). Filtered extractions were analyzed for a suite of major and trace elements using ICP-OES (Perkin Elmer Optima 3300 RL). The digestion procedure extracted > 80 % of both major and trace metals when compared to the certified reference values (GBW 07310) and precision was < 10 % RSD.

Triplicate sediment cores for pore water analysis were collected from the natural and de-embanked saltmarsh using PVC tubing and immediately sealed in the field to minimize oxidation. The samples were returned to the laboratory and stored at -20 °C until required. All sample handling was carried out in a N_2 atmosphere. Pore water was extracted using rhizon samplers at 2 cm intervals with the first rhizon port at 0.5 cm depth and frozen at -20 ºC until analysis. NH_4^+, NO_3^- and SO_4^{-2} were selected to indicate the extent of pore space connectivity with tidal floodwaters and the redox environment and were analyzed on a Skalar SAM++ segmented flow auto-analyzer.

3. Results

3.1. Broad-scale physical sediment characteristics

Average values for physical sediment characteristics (MC%, LOI% and dry BD) in natural and de-embanked saltmarshes are presented in Table 1 and demonstrate little variability in sediment characteristics across the study sites except for within recently de-embanked sites. Dry bulk density ranges from 0.26 to 1.47 g cm^3 with the highest values present for the de-embanked sites, whilst both MC (29-225 %) and LOI (5.7-29.6 %) are highest in the natural sites; however, there is only a significant difference for those sites de-embanked most recently through MR (Mann-Whitney U test, n=18, $p<0.05$). Figure 2 illustrates the difference between sediment characteristics in natural and restored sites (n=9, error bars are present, but very small) and this shows that although sediments in de-embanked sites may have characteristics which are different to their natural counterparts in the early stages of restoration (17 years for Tollesbury and Orplands, 21 years for Northey) this difference is not apparent for those sites de-embanked in the mid-20th Century and late 19th Century.

Table 1: Mean values for physical sediment characteristics for natural (N) saltmarshes and those de-embanked (DE) through managed realignment (MR) and storm surges in the 1950s and 1890s.

		Dry BD g cm^{-3}				Moisture Content (%)				LOI (%)			
		N		DE		N		DE		N		DE	
		mean	SD	mean	SD	mean	SD	mean	SD	mean	SD	mean	SD
MR	Tollesbury	0.29	0.02	1.40	0.12	221	3	33	5	28	1.2	7	1.2
	Orplands	0.55	0.06	1.06	0.03	126	9	50	6	16	1.5	9	0.6
	Northey	0.48	0.03	0.85	0.07	142	5	100	22	18	0.6	16	3.4
1950s	Barrow Hill	0.61	0.11	0.47	0.04	104	11	106	19	13	1.6	10	0.9
	Ferry Lane	0.73	0.0003	0.68	0.01	98	3	85	3	13	0.5	9	0.6
	Wallasea	0.59	0.03	0.61	0.05	72	6	88	6	9	0.2	11	0.9
1890s	Northey	0.48	0.03	0.69	0.12	142	5	94	16	18	0.6	13	2.3
	N Fambridge	0.60	0.04	0.58	0.08	81	13	102	6	9	1.4	10	0.4
	Brandy Hole	0.53	0.02	0.67	0.01	126	11	98	5	15	1.3	12	1.2

Sediment characteristics with depth were also examined at each site. Profiles with depth all show similar patterns and hence random representative profiles are given in Figure 3. In the natural sites and those de-embanked in the 1950s and 1890s BD shows little variation with depth, but increases rapidly with depth at all three MR sites. MC shows a very similar profile in both natural and de-embanked sites and is slightly elevated in the upper sediments decreasing with depth. However, for the MR sites these moisture contents are much lower. Similarly, LOI has a similar profile in all sediment cores, decreasing with depth; however in the MR cores, the LOI is much lower.

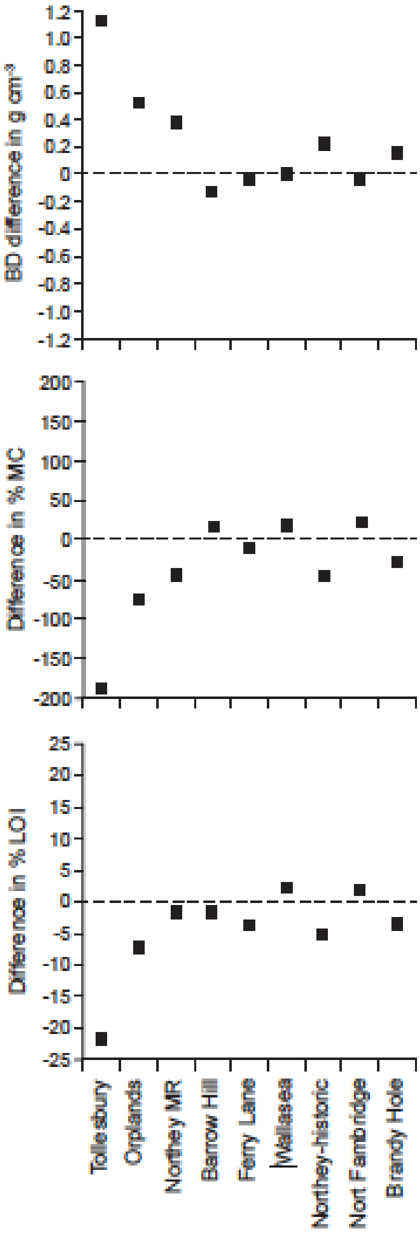
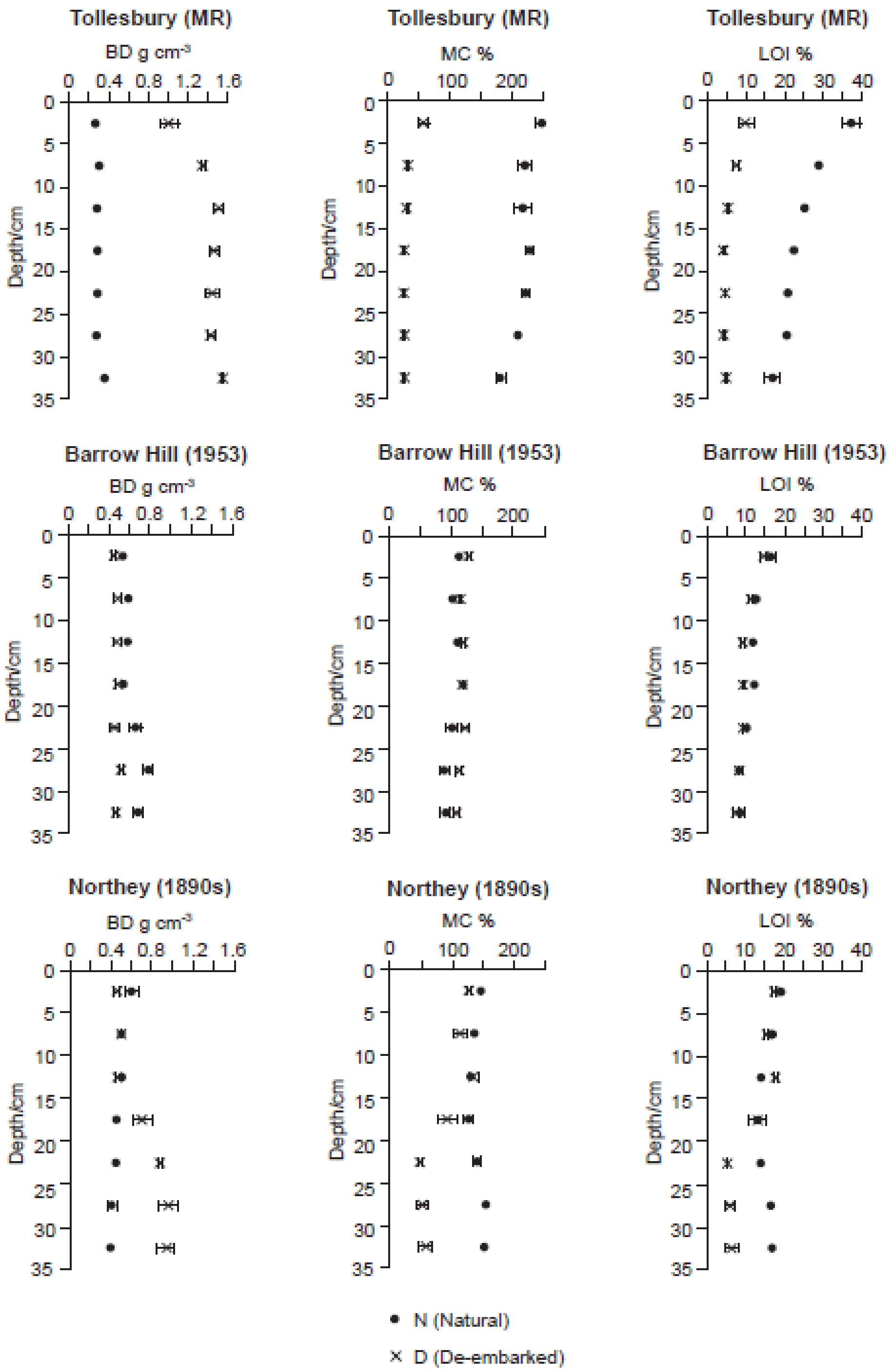


Figure 2: Difference between bulk density, moisture content and percentage loss on ignition for natural and de-embanked saltmarshes (mean data for each core +/- SE although not visible).

Figure 3: Bulk density, moisture content and percentage loss on ignition with depth in sediment cores from Tollesbury, Barrow Hill and Northey natural and de-embanked saltmarshes.



3.2. Intensive-scale physico-chemical sediment characteristics at Orplands Farm

Sediment structure – field observations

Sediments from the natural saltmarsh site comprise sandy-silts becoming more consolidated and drier below c. 35 cm depth. Organic material and occasional large macropores (> 1 mm diameter) are visible, whilst there are visible horizontal laminations throughout the cores. Sediments from the de-embanked MR site are also sandy-silts, becoming drier and firmer with depth. Two sediment facies can be identified in the R sediments; an upper sediment layer that is poorly consolidated with abundant root material and the presence of infrequent macro-pores and a lower, firmer sediment layer with less organic material, and patches of orange/brown sediment within the darker grey matrix indicating oxidation spots. There is a sharp, irregular boundary between these units varying from 2 - 8 cm depth below the saltmarsh surface.

Sediment structure - bulk phases

Representative reconstructed volumes of both natural and de-embanked sediment cores imaged using µCT are shown in the graphical abstract. These datasets identify four main bulk phases in the natural saltmarsh and five phases in the de-embanked saltmarsh. The dominant phase in all samples represents sandy-silts and can be sub-divided into three components; a lower attenuating phase (low density clay) found throughout the natural saltmarsh and in the upper sediment facies of the de-embanked saltmarsh (colored pink in the graphical abstract); and two progressively higher-attenuating phases found only in the lower sediment unit of the de-embanked cores (colored grey and brown respectively in the graphical abstract). It is likely that the mineralogical composition of sediments is similar as the underlying geology is the same and the sample locations have the same estuarine sediment source. Therefore, the differences in X-ray attenuation reflect the relative degree of microporosity at sub-voxel scales, resulting from a partial volume effect (Cnudde and Boone, 2013). Thus, we suggest the natural saltmarsh sediments and the upper facies of the de-embanked sediments have significantly higher microporosity than the lower sediment facies in the de-embanked cores (Table 2). The highest density component (brown in the graphical abstract) has a distinctive morphology and is interpreted to represent soil aggregates generated by the pre-breach agricultural land-use at the site.

Graphical Abstract: Sediment phases in A) natural and B) de-embanked saltmarsh cores.

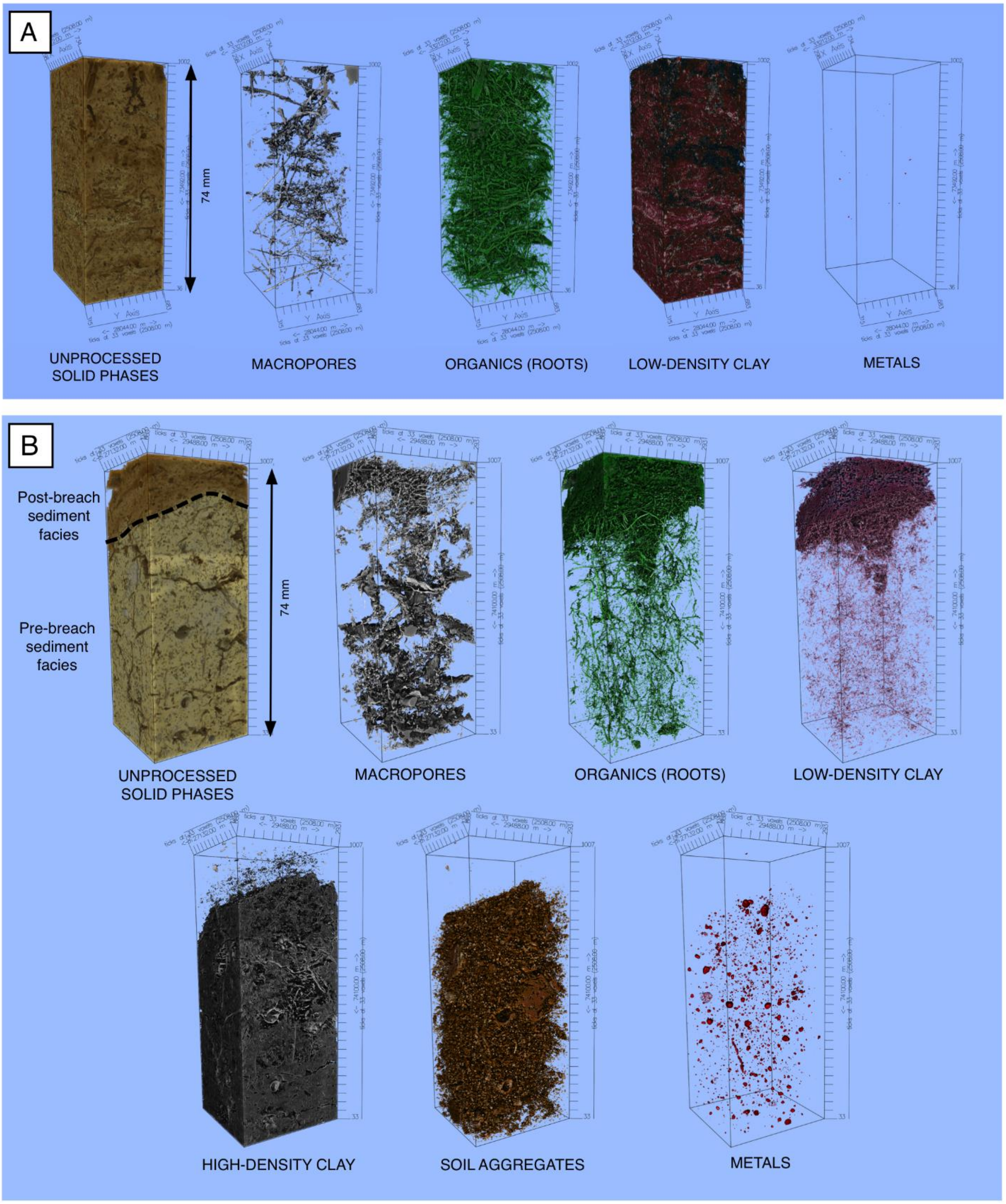


Table 2: Porosity analysis derived from µCT datasets

	Natural Saltmarsh A-F	De-Embanked Saltmarsh A-E (lower Facies: pre-breach)	De-Embanked Saltmarsh F (upper Facies: post-breach)
BULK POROSITY			
% Macroporosity	~3.6%	~9.8%	~8.3%
Microporosity	High: Greyscale value ~17500	Low: Greyscale value ~20000	Very High: Greyscale value ~16250
Macro-pore abundance	Low (mean 2273) but highly variable between samples	High (mean 5388) with less variance between samples	Low - Moderate (mean 2978)
Pore-Size Distribution	Variable, but abundance dominated by smallest pores (<0.01mm^3) Lower sub-samples tend to have smaller macro-pores, upper sub-samples have more variable in pore-size distribution.	Highly variable, with no trend of pore-size distributions.	Relatively few, but larger macro-pores. Also highest degree of pore size/volume variability.
Mean Pore Diameter	155-190 µm, upwards trend of increased variability.	250-700 µm, no distinctive patterns evident.	1831 µm, high variability between samples.
PORE NETWORK EFFICIENCY			
Pore Connectivity	Euler numbers moderate (~<2000) decrease upwards: increasingly simple, direct pore network, but variable.	High Euler numbers (~ -5000), weak decrease upwards in samples: very	Moderate to low Euler numbers (~ -1454). Simple, more direct pore network.

	Natural Saltmarsh A-F	De-Embanked Saltmarsh A-E (lower Facies: pre-breach)	De-Embanked Saltmarsh F (upper Facies: post-breach)
		interconnected, distributed and variable network	
Pore Anisotropy	Moderate to high, increasing upwards in all samples, but variable. Pores organized in a vertically aligned network.	Very low, random orientation of voids.	Very low, random orientation of voids.
Pore Network Size (no. branches per pore)	Low (~5000 - 11000 branches)	Very High (>25000 branches) and highly variable	Low to Moderate (~8000 - 14000 branches)
Pore Network Complexity	2.5-5 branches per pore	5-9 branches per pore	2-7 branches per pore

A second sediment phase present in both volumes is identified as organic matter showing characteristic rootlet structures (graphical abstract), although it is not possible to discriminate between live or decayed plant roots and other organic detritus. Abundance, distribution and orientation of roots and organic matter differs between the natural and de-embanked sediments. In the natural saltmarsh sediment organic matter is abundant and distributed throughout the sediment core (graphical abstract), whilst in the de-embanked sediments organic matter is less abundant, particularly in the lower sediment facies. Additionally, the dominant orientation of roots in the upper sediment facies of the de-embanked sediment is horizontal, whilst vertical roots dominate in the lower sediment facies.

Throughout both natural and de-embanked sediments, a third very attenuating (and hence higher density) bulk phase ('metals' and coloured red in graphical abstract) is recognized with two distinctive forms (graphical abstract). In the natural saltmarsh sediment this phase typically occurs as a coating around macro-pores and rootlets and probably represents iron plaques (Sundby *et al.*, 1998). This phase also exists in both the natural and the de-embanked sediments as discrete, small (<1 mm diameter) inclusions, but occurs in far greater abundance and with greater size in the lower sediment facies of the de-embanked sediments (graphical abstract). These probably represent iron and other metal precipitates. Finally, the lowest attenuating phase represents macropore space within these sediments and is analyzed separately below.

Sediment structure - macroporosity and pore networks

Mean values (n = 3) for macroporosity (pores > 80 µm; Beven and Germann, 2013) plotted against depth are presented in Figure 4, with a summary of qualitative and quantitative porosity measurements for each sediment facies given in Table 1. Bulk macroporosity is low (< 10 %) in both natural and de-embanked sediments. There are no consistent trends of macroporosity with depth, although mean values are considerably higher and more variable within the de-embanked saltmarsh, which is perhaps surprising given the generally higher measured BD in these samples (section 3.1). In the natural saltmarsh samples, macropore abundance and the number of discrete macropore networks are variable and do not display any trends with depth, but are consistently lower than that derived from both facies of the de-embanked samples. In the de-embanked samples macropore abundance is high and generally less variable in the lower facies, whilst in the upper facies abundance is lower and more comparable to that of the natural saltmarsh. In the natural saltmarsh samples, macroporosity is strongly skewed towards pores with volumes < 0.01 mm^3, whilst in the de-embanked sediment samples larger pores with volumes 0.01 - 0.1 mm^3 are also very common. Mean pore diameter offers an alternative measure of pore size, and in the natural saltmarsh samples is more consistent and varies between c. 150 – 190 µm whilst in the de-embanked sediments mean pore diameter is c. 250 – 700 µm in the pre-breach lower facies and 1831 µm in the post-breach upper facies.

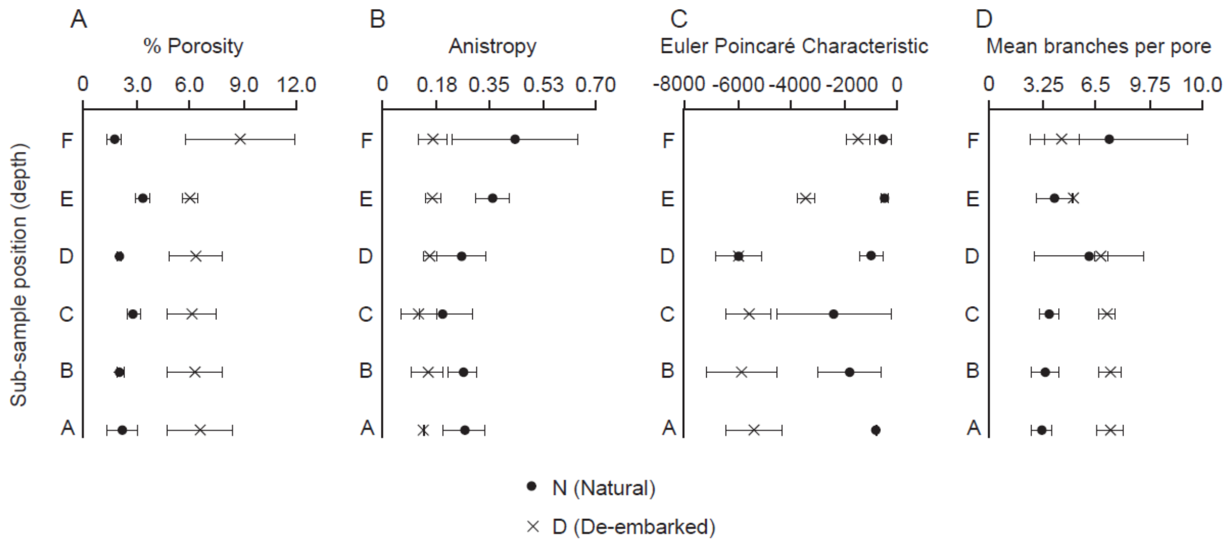


Figure 4: Porosity characteristics in vertical sediment cores from natural and de-embanked saltmarsh.

Examples of the 3D macropore network in both natural and de-embanked samples are illustrated in the graphical abstract, whilst mean values (n = 3) for the macro-pore network characteristics - connectivity and anisotropy with depth (six sub-samples A – F), are given in Figure 4 and Table 2. The Euler-Poincaré characteristic is a measure of the number of redundant connections within the pore network, and indicates the degree of connectivity and tortuosity with values closer to zero indicating fewer redundant connections and less tortuosity (Vogel, 1997). This value is typically expressed as a function of volume, but the raw data are presented here as the sub-samples are all of identical volume. Mean values for the Euler-Poincaré characteristics are moderate in the natural saltmarsh, but much higher in the de-embanked sediments suggesting that pore networks have more redundant connections and are more tortuous. The Euler-Poincaré characteristic is highly variable within cores, and does not show consistent trends with depth, however, there is a notable decrease in the upper sediment sub-sample (F) in the de-embanked sediments where values approach that of the natural saltmarsh. The degree of anisotropy of the dominant pore space is a reflection of how direct and similarly-arranged the branches of the dominant macro-pore system are (Odgaard, 1997). In the natural saltmarsh anisotropy is moderate to high with a vertical arrangement of the pore network, whilst in de-embanked sediment anisotropy is very low with no preferential arrangement of the pore network. Finally, the pores were transformed into topological networks of nodes and branches, permitting extraction of mean branches per pore (Figure 4), indicating the complexity of the macropore networks (Polder *et al*., 2010). There is a contrast in pore complexity indicated by mean number of branches per core where the de-embanked sediments (lower facies) display much greater complexity and variability than within the samples from the natural saltmarsh (Figure 4).

Sub-surface water levels

A detailed assessment of sub-surface hydrology for this study site, including a longer time-series analysis of water levels and event-scale hydrographs have been previously published (Tempest *et al.*, 2015). Mean water levels over two months for triplicate pressure transducers are shown in Figure S1. Patterns for water level fluctuations in both the natural and de-embanked saltmarsh are similar and reflect tidal flooding. However, in the de-embanked saltmarsh, water levels are higher and less responsive to tidal flooding. The similarity between the two times series is greatest during spring tides, when the surface of the saltmarsh is flooded. Differences between the triplicate times series may represent small-scale variations in micro-topography, and suggest that the natural saltmarsh is more heterogeneous than the de-embanked site.

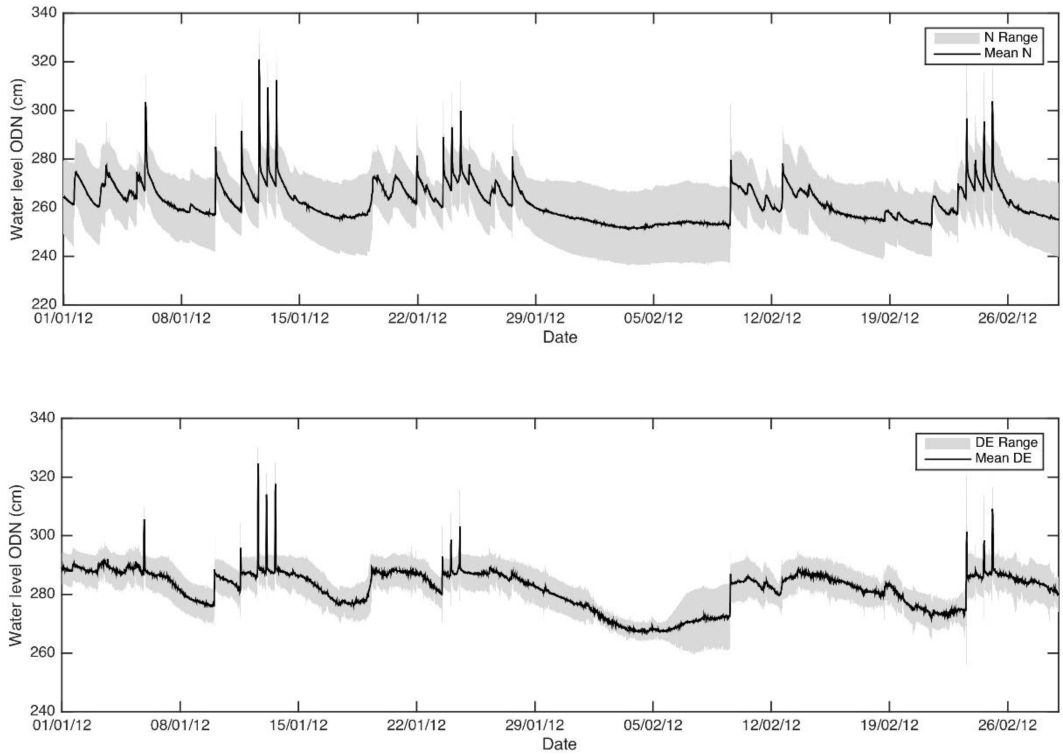


Figure S1: Average water levels from triplicate loggers positioned within the natural (a) and de-embanked (b) saltmarsh.

Sediment and pore water geochemistry

The sediment geochemistry has been explored in detail previously (Spencer et al., 2008) and here those elements indicative of tidal inputs (Na) and the sediment redox environment (Fe) are given in Figure 5. Sodium concentrations are higher in the natural saltmarsh and show a clear decrease with depth compared to the de-embanked saltmarsh, whilst concentrations of Fe are similar and show quite variable vertical profiles. Aluminium was used as a geochemical normalizer to compensate for grain-size differences in the sediment. Here, Al concentration varies with depth and suggests that sediment input varied between the two sites, probably as a result of the de-embankment process and different hydrodynamics between the sites. Figure 5 shows vertical profiles of Fe normalized to Al and indicates clear enrichment of Fe in the upper sediments of the natural saltmarsh that is not apparent in the de-embanked sediments.

Pore water concentrations of NH_4^+, NO_3^- and SO_4^{-2} with depth are shown in Figure 5. Pore water chemistry was highly variable between core replicates indicating heterogeneity in the sediment geochemical environment. Vertical profiles for NH_4^+, NO_3^- and SO_4^{-2} are quite typical for the natural saltmarsh cores with a decrease in NO_3^- and SO_4^{-2} with depth and an increase in NH_4^+. In contrast SO_4^{-2} and to a lesser extent NO_3^- increase with depth in the de-embanked saltmarsh and NH_4^+ concentrations peak with depth before decreasing. Both SO_4^{-2} and NH_4^+ concentrations are higher in the de-embanked saltmarsh.

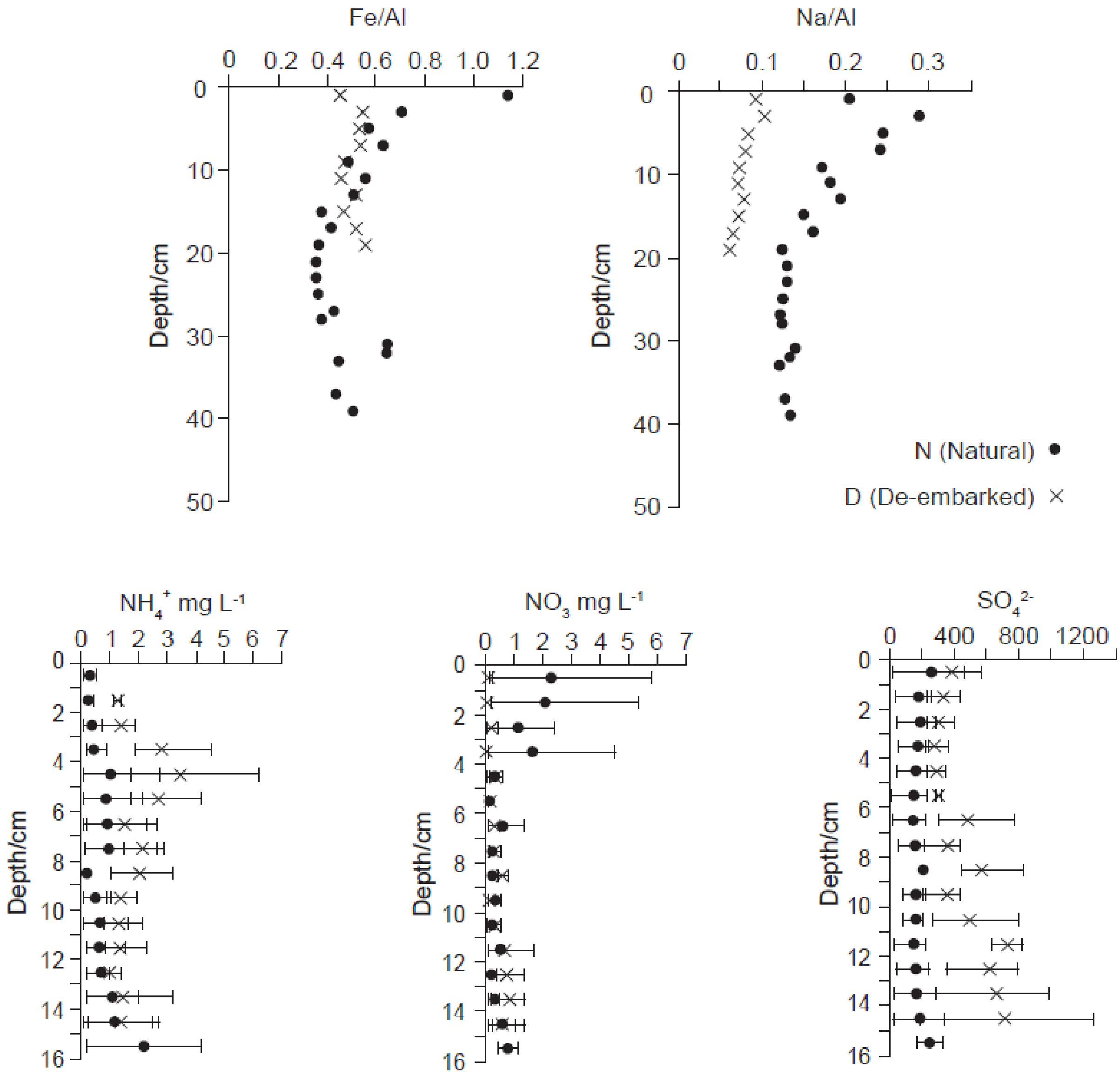


Figure 5: Sediment and pore water geochemical profiles with depth within the natural (a) and de-embanked (b) saltmarsh.

4. **Discussion**

4.1. Physical sediment characteristics and structure

The broad-scale investigation indicates there are clear differences in the overall sediment characteristics of natural and de-embanked sediments, most apparent and statistically significant for those sites de-embanked through MR. Natural saltmarsh sediments have lower bulk density, higher organic matter (LOI) and higher moisture content than the de-embanked saltmarshes. The natural saltmarsh shows typical vertical profiles with LOI and moisture content decreasing with depth indicative of the microbial breakdown of organic carbon and compaction (e.g. Spencer 2002). In contrast, sites restored through MR show two distinct sediment facies, with high bulk density sediments overlain by less consolidated sediment. These two sediment facies can clearly be observed using μCT (graphical abstract).

Prior to de-embankment, these sites were drained and used for agriculture, resulting in dewatering, mineralization of organic carbon, shrinkage of clays, and for the recent MR sites, compaction by modern agricultural machinery (Hazelden and Boorman, 2001; Boorman *et al.*, 2002; Ellis and Atherton 2003). Hence, this sub-surface horizon represents the pre-restoration 'relict' land surface and occurs at varying depths, arising predominantly from the topographic heterogeneity of the relict (ploughed) land-surface. Using μCT, the relict land surface is identified at c. 6 cm depth and is in broad agreement with previous studies (Spencer *et al.*, 2008). Assuming that sediment accretion has kept pace with regional relative sea level rise (3 mm a^{-1} see Woodworth *et al.*, 2009), the relict land surface should be at approximately 19 and 37 cm depth in the sites restored in the 1950s and 1890s respectively and is therefore below our sampling depth and/or not apparent in the older sites. Therefore, pre-restoration land-use has a significant impact on sediment characteristics and structure that persists for at least two decades following de-embankment.

4.2. Porosity

The application of μCT enables the detailed examination of 3D sediment structure allowing the estimation of relative microporosity and the quantification of macroporosity and pore networks. This determined that sediments in the natural and de-embanked saltmarsh differ in terms of microporosity, bulk macroporosity, pore morphology and pore connectivity.

The pre-restoration de-embanked sediment facies has lower microporosity than natural saltmarsh and this is supported by higher bulk density values in the broader-scale study in all the MR sites. Shrinkage of clay minerals following de-watering (Ellis and Atherton 2003) is likely to be exacerbated in these SE England sites, where the Ca-poor clays disperse (Crooks and Pye 2000) causing sediment fabric to collapse. This lowering of microporosity persists for at least several decades and is probably irreversible. As these sediments have mean bulk porosity < 10 % meaning they are likely to have low hydraulic conductivity and hence microporosity is particularly important for both storage and transport of water.

The pre-restoration de-embanked sediment facies has a higher mean bulk porosity, a higher abundance of individual macropores and larger diameter macropores than the natural saltmarsh. In the pre-restoration sediment facies pore network morphology (low anisotropy, redundant connections and more tortuosity) reflects a legacy of pre-restoration land-use through drainage, desiccation cracks and ploughing. Whilst, in the natural saltmarsh macropores largely result from roots and burrows, similar to the post-restoration sediment facies. Critically, the μCT data (graphical abstract) indicate low penetration of roots and burrows into the lower sediment facies, resulting in poor vertical connectivity of macropore networks. This implies that although the de-embanked sites overall have a higher macroporosity, the pore networks are inefficient at transporting water through the sub-surface environment.

4.3. Implications for sub-surface hydrology and connectivity with tidal floodwaters.

Water is stored in saltmarshes through either saturation storage resulting from the displacement of pore gases or dilation storage resulting from the swelling of clay minerals and the elasticity of organic matter (Knott *et al.*, 1987; Hemond *et al*., 1990). As a result, with changes in precipitation, saltmarsh volume and elevation vary seasonally (Cahoon *et al.*, 2006), and some studies have shown that saltmarshes swell following de-embankment as pore space fills with water (Paquette *et al.*, 2004; Anisfield 2012). However, in all three MR sites, mean moisture content remains low decades after restoration. This suggests that the changes to sediment structure noted above and low organic matter content (c. 4 - 5 % LOI in the sediment at depth) may impact the long-term flood storage capacity of restored saltmarshes.

Water flux and hence soil moisture conditions in tidal wetlands are controlled by a combination of the tidal regime, hydroperiod, topography (e.g. elevation and distance to creek networks), groundwater flow, precipitation and evapotranspiration (Hemond and Fifield 1982; Xin et al., 2013a and 2013b). This is further modified by sediment texture, flora/fauna (bio-irrigation, burrowing, roots) and stratigraphy (Gardner 2007; Wilson and Morris 2012; Xin *et al.*, 2009; 2012; 2013a; 2013b). In fine sediments, pore water movement is principally controlled by the microporosity and lateral movement is limited (Marani et al., 2006), although, macropores and sediment stratigraphy can also influence flow rates and pathways (e.g. Xin *et al.*, 2009).

In this study a time-series analysis of sub-surface water levels was used to indicate how efficiently water moves through the sub-surface environment (water level fluctuations and a detailed discussion of sub-surface hydrology are presented in Tempest *et al.*, 2015). The time series data (Figure S1) demonstrate that sub-surface hydrology in both the natural and de-embanked saltmarsh responds to tidal flooding but the more subdued response in the de-embanked sites suggests slower flow rates The sites are matched in terms of distance to creek networks and their proximity to each other means that tidal regime and meteorological conditions are the same and unlikely to account for these differences. At high tide, water level is influenced by surface infiltration of tidal floodwaters, with surface water percolating down through the sediment and exfiltrating via creeks and the marsh edge (Gardner 2005; Cao *et al.*, 2012). This can result in horizontal flows, efficient drainage and good soil aeration near to creek edges (e.g. Xin et al. 2013a). In this study sediments are fine-grained and cores were collected > 15 m from the creek edge and hence vertical flow should dominate (Marani et al. 2006). We used sediment Na concentrations as a proxy for salinity and hence vertical connectivity with tidal floodwaters. If tidal flow is not restricted, pore water salinities should quickly equilibrate with surface waters following de-embankment (Roman and Burdick 2012). However, Na concentrations are much lower in the MR site, suggesting that saline floodwater does not infiltrate very efficiently. This suggests low microporosity has a significant impact on vertical water flow in de-embanked sites and the extensive macropore network is largely ineffective with respects to solute transfer.

4.4. Implications for the sediment geochemical environment

The vertical profiles of sediment and pore water species within saltmarsh sediments are typically controlled by strong physicochemical gradients in Eh and pH and the microbially mediated reduction of O_2, NO_3^-, MnO_2, $Fe(OH)_3$ and SO_4^{2-} with burial (e.g. Spencer *et al.*, 2003; Koretsky *et al.*, 2005). Mobile reduced Fe^{3+} species migrate to the upper oxic zone and re-precipitate as oxyhydroxides, whilst pore water concentrations of NO_3^- and SO_4^{2-} decrease with depth. Typical profiles are observed in the natural saltmarsh sediments (Figure 5). This is further supported by the µCT data where a high density bulk sediment phase (graphical abstract) coating roots and burrows is suggestive of Fe-rich plaques (Mendelssohn *et al.*, 1995) and concretions suggest the rapid oxidation and precipitation of Fe in pore spaces (Sundby *et al.*, 1998). This is in agreement with the field observation of sediment oxidation spots, indicating a vertical redox gradient and vertical movement of Fe-rich pore waters through the sub-surface environment.

When inter-tidal sediments are drained for agriculture there is extensive precipitation of Fe oxyhydroxides and other Fe-rich minerals in the plough zone (Auxtero *et al.*, 1991; Violante *et al.*, 2003). Re-flooding with saline waters is expected to remobilize this Fe through the dissimilatory reduction of sulphate and re-distribution of dissolved Fe via the advective forcing of local hydrology (Burton *et al.*, 2011; Johnston *et al.*, 2011). However, surface enhancement of Fe is not observed in the de-embanked sediments and the high abundance of large, high-density aggregates (graphical abstract) in the pre-restoration sediment facies may be indicative of the presence of Fe-rich precipitates. Microbial reduction of Fe could be limited by the supply of sulphate. In these sediments sulphate is available (Figure 5) but not being utilized, suggesting that organic matter is unavailable or inaccessible to sulphate reducing bacteria. Labile organic matter is delivered to saltmarshes via trapping at the surface, through in situ biomass production and downward mixing via bioturbation (Koretsky *et al.*, 2005). In the de-embanked MR sediments organic matter (LOI) is very low and degraded (Santin *et al.*, 2009), whilst µCT indicates there is little bioturbation (graphical abstract). Therefore, the microbial reduction of Fe^{3+} is probably limited by organic matter availability. The lack of surface enhancement of Fe could also mean that abiotic conditions are unsuitable for Fe^{3+} reduction, perhaps indicated here by low moisture content and hence soil aeration (e.g. Kostka *et al*., 2002) or that these abundant oxyhydroxides represent a significant, stable crystalline pool of Fe. In addition, the physical mechanism for redistributing mobile Fe species i.e. advective pore water movement, may be absent. It is likely that a combination of these mechanisms are responsible for the geochemical profiles observed, however it is clear that the impeded water movement and changes to sediment structure discussed above are also impeding solute movement.

Pore water nitrate concentrations also show atypical profiles in the de-embanked sediment, although less emphasis is placed on their discussion due to large standard errors. Highly variable rates of denitrification and N_2O production were also observed by Blackwell *et al.* (2010) in de-embanked saltmarshes and attributed to the lack of connectivity between nitrate-rich floodwaters and denitrifying bacteria in pore space as a result of drainage and collapse of sediment structure and this is likely to be the cause of variable nitrate concentrations at depth.

4.5 Wider implications for coastal wetland restoration

The hydroperiod is generally considered the main controlling parameter that provides the physical framework for biological succession in restored saltmarshes; get this right and successful ecological restoration will follow. Hence, elevation is the key design criterion in saltmarsh restoration projects including managed realignment, controlled reduced tide and beneficial sediment re-charge schemes. However, this study demonstrates that a return to tidal inundation has not restored the physical (sediment structure or sub-surface hydrology) or chemical (sediment and pore water geochemistry) framework in these sites, despite elevation being matched to their natural counterparts. This may be critical to understanding why many restored saltmarshes have poor habitat heterogeneity, poor species diversity and abundance, and lack equivalent species composition.

In the literature, poor species diversity and abundance in restored sites is frequently attributed to 'water-logging', 'saturation' or anoxia (e.g. Mossman *et al.*, 2012). However, it is now clear that although sediments deposited following de-embankment have high moisture content, moisture content at depth in the root zone remains low for several decades. Most studies focus on the importance of soil aeration for plant growth, however it is possible that in these sites sediment saturation may fall below the wilting point limiting plant growth. This has been observed where hydraulic conductivity is low and evapotranspiration is high resulting in oxic conditions at depth (Xin et al. 2013a).

In the surface sediments, poor drainage may result in the build-up of potentially toxic dissolved species such as S^-, high salinity and anoxia with implications for seedling germination. In addition, the lack of vertical connectivity and impeded sub-surface water flow, will impact biogeochemical cycles driven by redox gradients with consequences for ecosystem services such as climate regulation, detoxification and contaminant storage. For example, Morris *et al.* (2014) suggested that poor drainage impacts mercury methylation potential in restored saltmarshes. This study also indicates that the pre-restoration land surface acts as a physical barrier to vertical root penetration (and perhaps burrowing invertebrates) and this may have consequences for species composition, as establishment of long vertical roots is important for the successful establishment of seedlings (Balke *et al.*, 2011).

Finally, although saltmarshes are naturally heterogeneous, key structural characteristics e.g. pore abundance, pore-size distribution, pore diameter and pore connectivity are significantly less variable in the MR sediments compared to their natural counterparts and this will reduce the micro-heterogeneity of redox conditions, biogeochemical cycling and habitat development.

5. Conclusions

This novel integration of 3D sediment structural information with sub-surface hydrological and geochemical data reveals detailed evidence of the impact of pre-restoration disturbance and land-use on the structure of restored saltmarshes. The findings have significant implications for the functioning, potential ecosystem service delivery and design of restoration schemes. These data have been placed in a wider context of other restored saltmarshes in SE England including sites naturally de-embanked due to storm surges in c. 1890s and the mid-20th Century and those deliberately restored through removal of sea defenses to allow tidal inundation.

In these sites, pre-restoration disturbance – drainage and agriculture - has had significant impacts on physical sediment characteristics, most significantly, a reduction in microporosity and pore network connectivity. These impacts persist for at least two decades and are likely to be irreversible due to depleted organic matter content and the presence of Ca-poor clays. A broader-scale investigation did not reveal evidence of such disturbance in saltmarshes de-embanked through historic storm surges. However, at all sites de-embanked via MR, the physical sediment characteristics (bulk density, LOI and moisture content) are indicative of such disturbance. Therefore, the fundamental structural fabric of restored saltmarsh sediments has been disturbed and this has implications for the wider structure and function of restored saltmarshes and the ecosystem services they deliver.

Reduced microporosity and pore connectivity have impeded drainage and sub-surface water flow, whilst there is limited interaction between the sub-surface environment and overlying floodwaters. This may reduce flood storage capacity, and has implications for the fate and transport of all solutes in the restored saltmarsh environment including nutrients, contaminants and greenhouse gases. Poor drainage and the build-up of toxic chemicals in surface sediments, low moisture content at depth, a pre-relict land-surface which inhibits vertical root penetration and low structural heterogeneity may all contribute to the poor species and habitat diversity seen in many restored saltmarshes. Therefore, whilst hydroperiod, a key structural control on ecological development in saltmarshes has been restored, *hydrological regime* and the geochemical framework have not. Designing and engineering restoration schemes that mimic hydrological regime may be challenging. However, this understanding of the impact of disturbance on structure and function goes some way towards managing expectations and understanding what ecosystem services can be achieved.

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Supplementary information

Figure S1: Water levels from triplicate loggers positioned within the natural (a) and de-embanked (b) saltmarsh (2 column).

Graphical Abstract: Sediment phases in A) natural and B) de-embanked saltmarsh cores (2 column)

Figure 1: Location of sampling sites in south east England (2 column).

Figure 2: Difference between bulk density, moisture content and percentage loss on ignition for natural and de-embanked saltmarshes (mean data for each core +/- SE although not visible) (1 column)

Figure 3: Bulk density, moisture content and percentage loss on ignition with depth in sediment cores from Tollesbury, Barrow Hill and Northey natural and de-embanked saltmarshes (2 column).

Figure 4: Porosity characteristics in vertical sediment cores from natural and de-embanked saltmarsh (1.5 column).

Figure 5: Sediment and pore water geochemical profiles with depth within the natural (a) and de-embanked (b) saltmarsh (2 column).