



The impact of drying on structure of sedimentary organic matter in wetlands: Probing with native and amended polycyclic aromatic hydrocarbons



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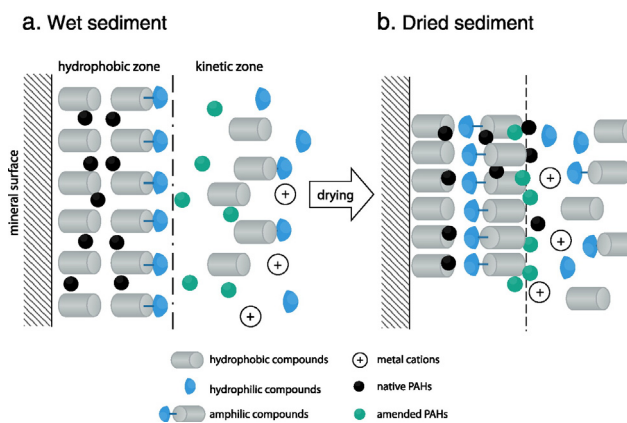
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HIGHLIGHTS

- Native PAHs were released into solution after sediments suffering dry-wet cycles.
- Drying may expose hydrophobic interior of SOM, where native PAHs are concentrated.
- Drying enhanced the fresh PAHs sequestration in the sediments.

GRAPHICAL ABSTRACT



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ABSTRACT

Wetland sediments undergo dry-wet cycles that may change their structural properties and affect geochemical behavior of associated organic compounds. In this study, we examined the effect of drying on particle size distributions and the rapid (24 h) sorption reactions of polycyclic aromatic hydrocarbons (PAHs) with salt marsh sediments in Nueces Delta, South Texas. Drying reduced the fraction of fine particles in organically richer sediments, indicating structural rearrangement of organic matter and mineral aggregates. Among the 16 EPA priority PAHs examined, dried sediment preferentially released 1.0–7.5% of phenanthrene, fluoranthene and pyrene to added seawater (solid: water mass ratio of 1/100) – significantly greater than release from sediments maintained in the wet state. On the other hand, drying also increased the affinity of sedimentary organic matter (SOM) for experimentally amended (deuterated) phenanthrene relative to continually wet sediments. Further, deuterated phenanthrene was even more effectively retained when it was added to wet sediment that was subsequently dried and rewetted. These apparently contradictory results can be reconciled and explained by SOM having a heterogeneous distribution of hydrophobic and hydrophilic zones – e.g., a zonal model. We propose that drying

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Zonal assemblage

changed the orientation of amphiphilic SOM, exposing hydrophobic zones and promoting the release of some of their native PAHs to water. Freshly amended PAHs were only able to penetrate into the surface hydrophobic zone and/or deeper but rapidly accessible (“kinetic”) zone in wet sediments due to the brief adsorption contact time. Subsequent drying presumably then induced structural changes in SOM that isolated these amended PAHs in sites inaccessible to water exchange in the next rewetting. These results provide insights into structural changes of SOM upon drying, and help predict the fate of compounds such as organic contaminants during drought/flood oscillations.

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1. Introduction

Soils and wetland sediments are often subjected to dry-wet cycles in nature, and these cycles may change the structural properties of organo-mineral complexes and affect the distribution of associated chemicals (e.g., Horne and McIntosh, 2000; Liu and Lee, 2006). Soil organic matter becomes fragmented and more porous after being dried (White et al., 1998). Drying may also affect the orientation of non-polar and polar moieties of soil organic matter on mineral surfaces and change its apparent surface and/or bulk hydrophobicity (Doerr et al., 2000; Liu and Lee, 2007). Such behavior is consistent with proposed models of organic matter in soils or sediments that involve hydrophobic interiors and more hydrophilic exteriors – e.g., so-called “micelle” structures (Wershaw, 1993, 2004; Sutton and Sposito, 2005). Thus drying-rewetting can affect the partitioning of organic contaminants between water and solid phases in aquatic environments (Liu and Lee, 2006).

Understanding structural changes of SOM is key to deciphering association of organic contaminants, yet it is difficult to directly investigate the SOM structure due to limitations of available techniques. For example, electron microscopy can detect surface features of SOM, but current versions cannot probe the inner structure of solid samples and, furthermore, often require dried samples in vacuum (Ghosh et al., 2000; Arnarson and Keil, 2001; Liu and Webster, 2007). Because structural changes affect sorption behavior of various compounds onto SOM, these sorption reactions provide probes of 3-dimensional (3D) structural changes. For example, drying increases release of some SOM-sorbed compounds into solution (Liu et al., 2008). As an alternative, different types of amended sorbates can be used as probes to explore structure of SOM (White et al., 1998; Kottler et al., 2001; Liu and Lee, 2006, 2007). PAHs, as a group of hydrophobic organic compounds, are excellent sorbates due to their high affinity to SOM and widespread existence in natural environments (Wang et al., 2014).

Studies have shown that drying-rewetting increases the sequestration of organic contaminants in SOM, indicating that drying can distort pore networks and/or the hydrophobic zone of SOM, where organic contaminants such as PAHs are typically sorbed (Shelton et al., 1995; White et al., 1998). Most studies used amended PAHs and dried sediments (e.g., Chiou et al., 1998; Kottler et al., 2001; Wu and Sun, 2010), but how drying affects native, or aged, PAHs in sediments remains unclear. The comparison between partitioning of freshly amended vs. native PAHs, – i.e. those that have been in contact with SOM for longer time scales – can provide extra insight into interactions of PAHs with SOM. It also can provide tests of conceptual models of the structure of organo-mineral complexes, such as the one proposed by Kleber et al. (2007).

Using PAHs as the probe and drying as an experimental treatment, we address two questions in this study: (1) how does drying affect partitioning of native and amended PAHs between solid and solution phases, and (2) what are the implications for SOM structure from the PAH sorption reactions in dried or wet sediments? These questions are important, considering the widely-distributed PAHs in aquatic environments and the impacts of projected future increase of frequency and intensity of droughts/floods (Zakaria et al., 2002; Huntington, 2006). By comparing partition behaviors of both native and amended PAHs in

marsh sediments, we demonstrate that drying causes structural changes of SOM and further affects distribution of organic contaminants.

2. Materials and methods

We used laboratory batch experiments to examine effects of drying-rewetting on the sorption/release of PAHs from salt marsh sediments, and to gain insights into structural properties of SOM. We studied the release of native PAHs from wet and dried sediments having different physical and chemical properties such as grain size distribution and total organic carbon contents. The release of native PAHs from different size fractions of marsh sediment was also compared, with the aim of testing the effects of grain size and associated SOM on the PAH release. We further tested the impact of drying on SOM, sorption and desorption of amended phenanthrene (deuterated Phe- d_{10}) among wet, dried, and dried/rewetted sediments.

2.1. Sample collection and pretreatment

We sampled salt marshes of the Nueces River Delta, South Texas (Fig. 1). Sediments in this area consist of mainly quartz (>90%), plus a small fraction of clay minerals including montmorillonite, illite and kaolinite in decreasing abundance order (White et al., 1983). Surface sediments (top 5 cm) were sampled during June–October 2011 using home-made core tubes. The sampling stations covered different types of wetland environment previously studied in this area (Dunton et al., 2001; Liu et al., 2013), including Nueces River riverbed at station LP (STLP), Nueces River mouth at station 104 (ST104), Nueces Bay at station 80 (ST80), and Nueces marsh at stations 450 and 451 (ST450, ST451). All cores were capped and stored in a freezer at -20°C before processing. A portion of surface sediment (0–5 cm) was dried in an oven at 40°C for at least 24 h until the sediment weight became constant. Drying at temperatures such as 40°C has been shown to lead to inconsistent changes in water repellency, which is related to exposure of hydrophobic sites (Dekker et al., 1998; Doerr et al., 2000). However, little difference in lysine sorption was found between marsh sediments dried at 60°C in the oven vs. a freeze-drier (Liu and Lee, 2006), indicating that different ways of drying do not significantly change sorptive properties of the SOM.

Portions of surface sediment were kept in a -20°C freezer without drying. Seawater for making batch slurries was collected at each station and filtered through pre-combusted $0.7\ \mu\text{m}$ GF/F filters (Whatman). Background concentrations of PAHs (total extractable PAHs) in seawater and sediments (the latter are termed “native” or “aged”) were measured for each station (Fig. A1 in Appendix, Table 1).

2.2. Sediment characteristics

Black carbon (BC) contents in sediments were measured according to Gustafsson et al. (1997). Briefly, inorganic carbonates (IC) in dried sediments were removed by acid fuming in a sealed container with concentrated hydrochloric acid (Hedges and Stern, 1984). One fraction of the acidified sediment was measured by an elemental analyzer to obtain total organic carbon (TOC) content. The other fraction was placed in a

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