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Distribution, input pathway and mass inventory of black carbon in sediments of the Gulf of Thailand, SE Asia

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ABSTRACT

The coastal margins around Southeast Asia (SE Asia) may serve as an ideal location to study the source-sink process of sedimentary black carbon (BC) because SE Asia has been identified as one of the major BC emission source regions in the world. This study provides an extensive picture of recent regional-scale sedimentary BC sequestration in the Gulf of Thailand (GOT), a tropical marine system in SE Asia. Generally, the sedimentary BC concentrations (0.07–3.99 mg/g) were in the low to moderate ranges of those obtained in other coastal sediments around the world. Regional variability of the BC and its correlation with the sediment grain size and total organic carbon (TOC) content indicated a general hydrodynamic constraint on BC occurrence in the lower Gulf in contrast to the upper Gulf with a more source dependence due to the direct land-based input. BC/TOC% values and the varied BC components (char and soot), as well as their correlations suggested that char was the predominant constituents of sedimentary BC both in the upper and lower Gulf, which could be mainly derived from biomass burning and entered into the nearshore region through direct fluvial transport and surface run-off. The estimated BC burial flux (~212 μg/cm^2/y) and mass inventory (~200 Gg/y) in the GOT on the hundred-year timescale were of the same order of magnitude compared with other oceanic margins, and thus the tropical shelf sediments from SE Asia could serve as an important sink of land-emitted BC.

1. Introduction

Black carbon (BC), including both the char and soot components, is usually defined as the recalcitrant carbonaceous residue produced from the incomplete combustion of biomass and fossil fuels (Goldberg, 1985; Schmidt and Noack, 2000; Baldock and Smernik, 2002; Dickens et al., 2004). The widespread combustion process and refractory nature make BC widely distributed in environmental matrices, such as the atmosphere, soils, fresh/sea water, ice and sediments. The emitted BC can eventually be transferred into the ocean through direct atmospheric deposition and river run-off by soil erosion (*e.g.*, Goldberg, 1985; Suman et al., 1997; Gustafsson and Gschwend, 1998; Kuhlbusch, 1998; Mitra et al., 2002; Elmquist et al., 2008; Lohmann et al., 2009), which is an important process of the global carbon cycle as transferring the carbon from a rapidly cycling atmosphere-biosphere system into

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the long-term geological carbon pool (Seiler and Crutzen, 1980; Suman et al., 1997; Kuhlbusch, 1998; Middelburg et al., 1999). The interaction of BC among the various carbon pools is also relevant in predicting the climate change of the future earth (Bond et al., 2013).

It has been estimated that ~90% of the marine BC deposition occurs on the marginal settings with the area less than 10% of the world ocean (Suman et al., 1997); however, the magnitude of the BC mass inventory in marine sediments on the global scale remains largely unknown (e.g., Gustafsson and Gschwend, 1998; Dickens et al., 2004; Masiello, 2004; Elmquist et al., 2008), which may be partially due to the scarcity of large-scale observations. Therefore, the extensive study of the BC occurrence and mass inventory in the continental margins is essential for a better understanding of the global biogeochemical cycle of BC. However, a vast majority of the existing studies on the sedimentary BC in the coastal margins were geographically within high-latitude regions (*e.g.,* Europe and America), such as the Northern European shelf (Sánchez-García et al., 2012), the Gulf of Cádiz (Sánchez-García et al., 2013), the Gulf of Maine (Gustafsson and Gschwend, 1998; Flores-Cervantes et al., 2009), the pan-arctic estuaries (Guo et al., 2004; Elmquist et al., 2008); the Washington coast (Dickens et al., 2004), etc., however, only little attention has been paid to the continental shelf of Asia (*e.g.,* Wang and Li, 2007; Sun et al., 2008), particularly for the Southeast Asia (SE Asia), a tropical region, which has been identified as a major BC emission source region due to frequent forest fires, biomass burning and escalating fossil fuel utilisation (Streets et al., 2003). Moreover, the tropical coastal margins in SE Asia also serve as an important source emission region for coastal BC export due to the inherent nature of rapid transferring of land-based materials into the aquatic system *via* strong rain and surface runoff (Nittrouer et al., 1995; Zakaria et al., 2002; Saha et al., 2009). Therefore, the coastal sediments from tropical SE Asia may be an important reservoir for the exported BC from the adjacent landmass. Nevertheless, to date, the characterisation of sedimentary BC in these tropical regimes remains poorly understood.

The Gulf of Thailand (GOT), a shallow and semi-enclosed shelf sea in SE Asia, has been subjected to a huge BC emission from biomass burning, forest fires and fossil fuel consumption from the adjacent regions (Saha et al., 2009; Sahu et al., 2011; Huang et al., 2013). Because of the nutrient-rich, shallow waters and their confined nature, the ecosystem of the GOT is especially vulnerable to human activities (Srisuksawad et al., 1997), and increased anthropogenic activities around the GOT have induced severe environmental pollution, especially for the upper Gulf with significant river export of land-based contaminants (*e.g.,* the Chao Phraya River) (Wattayakorn et al., 1998). Petroleum hydrocarbon contamination is evident in the nearshore waters of the GOT (Wattayakorn et al., 1998; Wattayakorn, 2012), and sedimentary polycyclic aromatic hydrocarbons (PAHs) exhibit a dominant river influence with mixed pyrogenic and petrogenic origins in the upper Gulf but with a dominant pyrogenic signature in the lower Gulf through atmospheric deposition (Boonyatumanond et al., 2006). This result may imply a different input pathway and source-sink processes for the combustion-derived substances (such as BC and PAHs) in the different GOT compartments (*e.g.,* the upper vs. the lower Gulf).

In the present study, the large-scale occurrence of sedimentary BC and its mass inventory in the GOT was first examined to present a comprehensive study on BC abundance and its distribution pattern, input pathway and mass inventory in this tropical marginal system.

2. Materials and methods

2.1. Study area and sediment sampling

The Gulf of Thailand (GOT) is a shallow, semi-enclosed tropical marine embayment situated in the South China Sea, which is surrounded by the land mass of Malaysia, Thailand, Cambodia and Vietnam (Fig. 1). The GOT is relatively shallow with a mean depth of 45 m and a maximum depth of 80 m. The Gulf can be usually divided into two parts, the upper Gulf and the lower Gulf (Fig. 1). The upper Gulf is the northernmost part of the GOT, covering approximately 10,000 km^2 and receives a large amount of sewage, runoff and sediments, especially from the Chao Phraya River, which has a catchment area of 162,000 km^2 and flows through several cities, including the mega-city of Bangkok (Wattayakorn et al., 1998; Boonyatumanond et al., 2006). The regional climate is generally warm and humid, with a typical southwest monsoon that prevails from May to September and a northeast monsoon that prevails from November to February. The induced seasonal circulation in the GOT is generally weak and variable (Wattayakorn et al., 1998; Wattayakorn, 2012), where an apparent counter-clockwise circulation occurs in the upper Gulf during the northeast monsoon, whereas both clockwise and counter-clockwise circulations occur during the southwest monsoon, depending on the wind conditions and external flow through the open boundary (Sojisuporn and Putikiatikajorn, 1998; Wattayakorn et al., 1998). The Gulf is poorly flushed on the whole, with little mixing, especially in the upper part (Wattayakorn et al., 1998), resulting in the majority of sediments and associated contaminants from the fluvial inputs primarily depositing in the upper Gulf (Srisuksawad et al., 1997). Sediment accumulation in the open GOT is mainly constrained by a combination of wind-driven currents, tides and its bottom topography (Takahashi et al., 1984). Muddy sediments mainly occur in the coastal area near the mouths of the major rivers in the upper Gulf, in addition to the central and western parts (near Samui Island) in the lower Gulf (Windom et al., 1982).

Ninety-three surface sediment samples were strategically collected on a regional-scale through three cruises conducted by *R/V* Boon-Lerd Pa-Sook and *R/V* SEAFDEC 2 (SEAFDEC, *i.e.,* Southeast Asian Fisheries Development Center) during 2010–2012. The samples were collected using a stainless steel box corer deployed from the vessels. All sediment samples (0–3 cm) were wrapped in pre-combusted aluminium foil and stored at −20 °C until analysis.

2.2. Analytical procedure

Due to its complex organic and mineral matrices, the separation and quantification of BC in sediments is complicated, and several procedures have been developed for specific studies (Gustafsson et al., 1997, 2001; Song et al., 2002; Hammes et al., 2007; Han et al., 2007a, 2007b; Khan et al., 2009; Poot et al., 2009; Meredith et al., 2012; Wiedemeier et al., 2015). In the present work, the wet-chemical treatment combined with thermal/optical reflectance (TOR) detection was adopted (Han et al., 2007a, 2007b; Fang et al., 2015). Briefly, the thawed, freeze-dried and homogenized (<80 meshes) sediment samples were treated with hydrochloric and hydrofluoric acids to remove inorganic materials. The acid-treated sediment residue was then filtered through pre-combusted quartz fibre filters (Whatman, 450 °C for 4 h and 47 mm in diameter). The filter samples were air dried and subsequently analysed for BC on a Desert Research Institute (DRI) Model 2001 Thermal/Optical Carbon Analyser (Atmoslytic Inc., Calabasas, CA) following the Interagency Monitoring of Protected Visual Environment (IMPROVE) protocols. A 0.544 cm^2 circular punch drilled from the filter was placed in a quartz boat and then sent into an oven. During the carbon analysis, the oven was first heated in 100% He atmosphere, producing four OC fractions (OC1, OC2, OC3, and OC4) in four temperature steps (140, 280, 480, and 580 °C). Then, the analysis atmosphere was shifted to 2% O_2/98% He, and three EC fractions (EC1, EC2, and EC3) were produced in three