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Baseline

Risk assessment and seasonal variation of heavy metals in settling particulate matter (SPM) from a typical southern Chinese mariculture base

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ABSTRACT

Settling particulate matter (SPM) samples were collected monthly during a 21-month-long (April 2014 to December 2015) monitoring program from a fish cage and a large seaweed (*Gracilaria lemaneiformis*) cultivation in a typical mariculture base in eastern coast of Guangdong Province, South China. The concentrations of eight heavy metals (Pb, Cr, Ni, Cu, Zn, Co, V and Mn) showed a clear seasonality with relatively higher levels in the rainy season for most metals. In addition, five metals (Pb, Cr, Ni, Cu and Zn) showed slightly higher concentrations in the fish cage than in the large seaweed cultivation. A principal component analysis (PCA) further revealed that Ni, Cu and Zn were influenced by human activities, whereas the rest of the metals mainly by a combination of natural and anthropogenic factors. SPM in the two different mariculture areas had a 21% probability of toxicity based on the mean effects range-median quotient.

Heavy metals, which usually involve > 40 kinds of metals with a specific gravity of 5.0 or over, have attracted much research attention worldwide for decades due to their toxicity, non-degradability and widespread pollution (Gu et al., 2012; Gupta et al., 2016; Zhuang et al., 2016). Coastal and estuarine ecosystems are important reservoir for many persistent pollutants such as heavy metals (Gao et al., 2014; Alyazichi et al., 2015; Gu and Lin, 2016). These metals enter estuaries and coastal waters via industrial and municipal wastewater discharges, storm run-off, dust deposition, mine discharge, waste incineration, and other diffusive processes (Matthai et al., 2002; Pan and Wang, 2012; Zhang et al., 2012; Zheng et al., 2013; Gascón Díez et al., 2016), and become biologically active when complexing with organic ligands (Callender, 2014).

Settling particle matter (SPM) is an important medium in the enrichment of heavy metals and subsequent removal from the water column to the sediment sink (Bruland et al., 2014). It is usually loaded with heavy metals due to its enrichment of metal-binding organic ligands, and mainly transported as both inorganic and organic suspended particles and deposited along the coast, leading to disruption in the delicate food web and significantly impacting the health of marine ecosystem (Feely et al., 1986; Zhang, 1999; Yiğiterhan et al., 2011; Helali et al., 2016). Therefore, SPM plays a critical role in the pollutant cycling and aquatic food chains in the aquatic systems (Jeremiason et al., 1998; Puig et al., 1999; Zhang, 1999; Gascón Díez et al., 2016). Despite of intensive study of metals in a variety of environments (Lofts, 2007; Qishlaqi et al., 2009; Sundaray et al., 2011; Yi et al., 2011; Gascón Díez et al., 2016), investigation of metals in SPM from food-producing aquaculture zones has remained scarce.

China has the world's largest aquaculture industry. Its aquaculture production accounts for ~72% of its reported domestic fish production and contributes > 60% of the global aquaculture volume and roughly half of the global aquaculture value (Cao et al., 2015). Since China plays a leading and decisive role in promoting aquaculture development worldwide and remains the world's largest exporter of fishery products (Cao et al., 2015; Gu and Lin, 2016), its production environment is critical to the quality of the aquatic products and food safety.

SPM contains numerous metal-binding organic ligands that can form chelate complexes with metals. The chelation process significantly increases the solubility of metals in the seawater (Bruland et al., 2014), thereby raising their biological availability and toxic potential to aquaculture organisms. Therefore, metals in SPM are closely related to the quality of aquatic products, and ultimately human consumers' food safety and health, highlighting the need for adequate evaluation of the metals concentration of SPM in aquaculture zone. The present study was designed to (1) survey the seasonal variations of SPM metal concentrations in two typical mariculture areas in South China, and (2) explore the sources and potential biological risk of the SPM metals.

Baisha Bay is located on the northeast coasts of Nan'ao Island,

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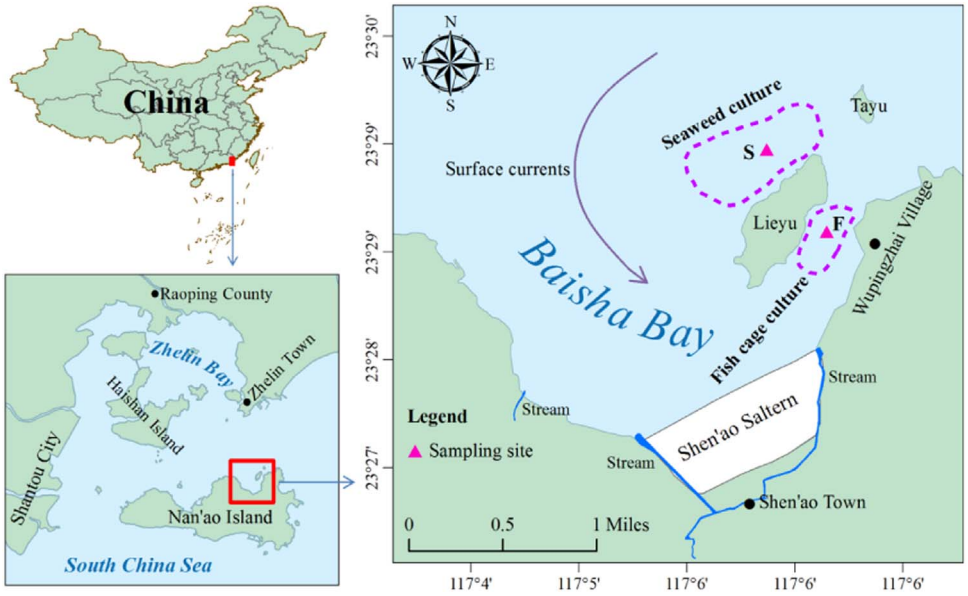

Fig. 1. Map of the Baisha Bay in South China Sea and the locations of settling particulate matter sampling sites. The surface currents based on Gu (2012).

Guangdong Province, South China (Fig. 1). It covers an area of 6.84 km^2 and an average water depth of 4.3 m with a maximum depth of 10 m. There are three streams discharging to this bay (Fig. 1). In this bay the mariculture industry has been developed since the 1980s and its mariculture zone is situated in a relatively sheltered shallow area (mean depth $<$ 3 m) including fish cage and large seaweed (*Gracilaria lemaneiformis*) culture areas. The fish-cage area is about 1 km^2 and has housed more than one thousand of 3 m $\times$ 3 m cages for over 30 years. The cultivation of large seaweed *G. lemaneiformis* at industrial scales in South China was established in 2000 along the coast of Nan'ao Island, and the farmed area dramatically increased from 0.13 ha in 2000 to 1500 ha in 2011 (Yang et al., 2015).

Two different types of mariculture areas in the Baisha Bay (Fig. 1) were selected to collect SPM samples using our self-designed sediment trap and the heights from seafloor of sediment trap in two different stations are same (Fig. 2). During the 21-month-long monitoring from April 2014 to December 2015, the SPM samples were collected once a month, and then placed in zip-lock polyethylene bags. All samples were preserved with ice and immediately transferred to the laboratory and kept at -4 °C until further analysis. Samples were freeze-dried to constant weight, then ground gently with agate pestle and mortar, sieved with 63 μm mesh sieve for homogenization, and then stored in glass bottles for metal analyses. These procedures eliminate variables such as moisture and grain size that are known to affect X-Ray Fluorescence (XRF) accuracy (Argyraki et al., 1997; Schwarz et al., 2012).

The concentrations of metals were measured using a Skyray EDX6000B XRF spectroscopy, a fast, nondestructive method proven to be comparable to Atomic Absorption Spectrophotometer (Muohi et al., 2003; Schwarz et al., 2012; Weindorf et al., 2013). Pressed powder tablets were prepared from finely powdered samples ($<$ 63 μm) following the procedures for direct elemental determination of the sample (Basta and McGowen, 2004; Verma et al., 2011). To verify the accuracy of the procedure, a China National Standard material (Offshore Marine sediment, GBW 07314) was applied to verify the XRF recovery and recoveries for the eight metals fell between 97% and 108%.

The monthly record of the heavy metal concentrations in SPM from the fish cage and large seaweed cultivation areas shows significant seasonality (Figs. 3 & 4), and the seasonal variation pattern was similar in these two different mariculture types. For the fish cage area, the peak values of Pb, Cr, Co and Mn were observed in August 2014, Cr and Ni in May 2014, Cu in November 2014, Zn in December 2014 and V in September 2015. For the large seaweed cultivation area, Pb, Co and Mn showed peak values in September 2015, Ni and V in June 2014, Cr in May 2014, Cu in November 2015, Zn in July 2014. Coincidently, the highest concentrations of all metals were founded between April–September of 2014 and 2015, partly owing to the more intense anthropogenic activities during the summer seasons. Moreover, the 2014 and 2015 summers were flooding season under the influence of monsoon climate, April–September 2014 and 2015 and the resulting dilution effect may also have caused the consequent reduction in the

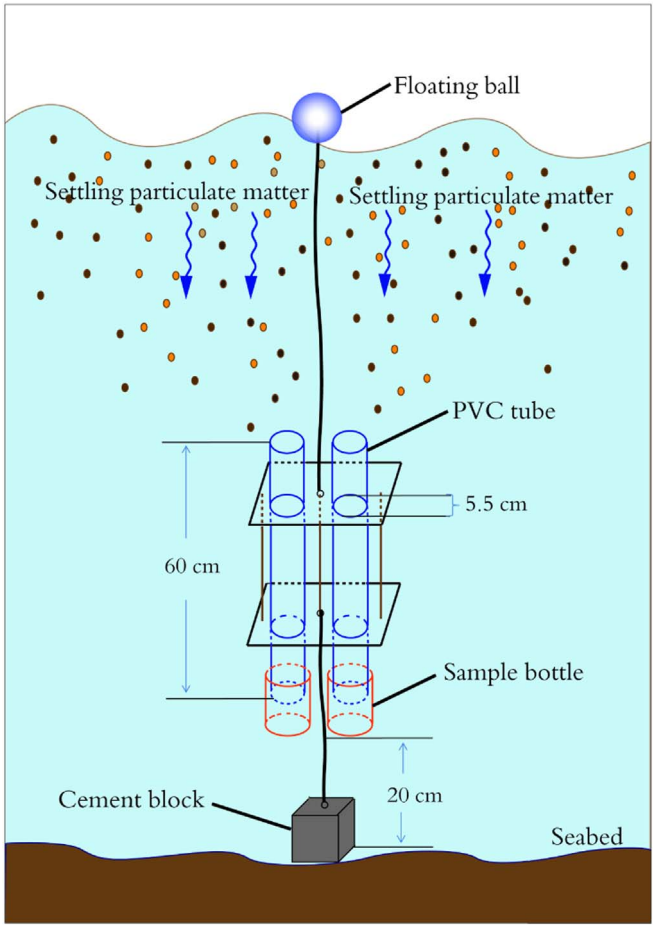

Fig. 2. Sediment trap designs for collecting settling particle matter.