Marine Pollution Bulletin 119 (2017) 365–371

Contents lists available at ScienceDirect

Marine Pollution Bulletin

journal homepage: www.elsevier.com/locate/marpolbul

Source apportionment of sediment organic material in a semi-enclosed sea using Bayesian isotopic mixing model

Jing Yu[a,b], Hua Zhang[a,*]

[a] Key Laboratory of Coastal Zone Environmental Processes and Ecological Remediation, Yantai Institute of Coastal Zone Research, Chinese Academy of Sciences, Yantai 264003,China

[b] University of Chinese Academy of Sciences, Beijing 100049, China

ARTICLE INFO

Keywords:
Stable isotopes
MixSIR Carbon budget
Atmospheric deposition
Particulate organic carbon

ABSTRACT

To determine sources of organic material in semi-enclosed Bohai Sea, samples of marine surface sediments, suspended particulates in adjacent rivers and atmospheric deposition were collected and analyzed for grain size composition, total organic carbon(TOC and POC), total nitrogen (TN and PN), and stable isotopic composition ($\delta^{13}C$ and $\delta^{15}N$). Measured bulk C/N ratio (5.50–12.28), $\delta^{13}C$ ($-23.59 \sim -19.54‰$), and $\delta^{15}N$ (2.80–8.07‰) values of surface sediment organic materials indicated a mixed source of marine and terrestrial contributions. Spatial distribution of organic C, N and their stable isotope composition indicated a land-sea gradient of organic material content and source combination. Using MixMIR model with dual isotopes, it was estimated that relative contributions of marine, riverine, and atmospheric sources to sediment mixture were 69.0%, 9.6%, and 21.4%, respectively. Our results demonstrated the advantage of Bayesian isotope mixing models over the conventional end-member mixing models for source apportionment in coastal seas with complex source origins.

1. Introduction

Marine sediment is the largest global reservoir in the carbon cycle [e.g., (Ramaswamy et al., 2008; Walsh, 1991)]; 80–90% of global organic carbon sinks via water column and buries in continental margin, though the area accounts for only 10% of the world ocean (Goni et al., 1997; Winkelmann and Knies, 2005). As the main interfaces between continent and ocean, marginal seas store significant amounts of sediment organic material (OM) from autochthonous (e.g., marine phytoplankton) or allochthonous sources (e.g., organic material from river, and carbonaceous materials from atmospheric deposition) (Sarkar et al., 2015; Serna et al., 2010) due to high sedimentation rates and biological productivity (Yao et al., 2015). Allochthonous contribution of OM through river discharge and atmospheric deposition is particularly important in semi-enclosed marginal seas with substantial terrestrial influences (Goni et al., 1997; Sarma et al., 2014; Usui et al., 2006). Recently, atmospheric transport of carbonaceous materials derived from incomplete combustion of biomass and fossil fuels received increasing concern in marginal seas downstream of wind from mainland (Fang et al., 2015; Zong et al., 2015). However, contribution of atmospheric deposition to marine sediment organic material remained unknown and needed to be quantitatively evaluated (Mahowald, 2011).

Carbon and nitrogen stable isotopic compositions ($\delta^{13}C$ and $\delta^{15}N$) and their elemental ratios (C/N) have been widely used as tracers to quantitatively elucidate the fate and origins of OM in the marine environments [e.g., (Liu et al., 2006; Thornton and McManus, 1994; Wada et al., 1987)]. Several studies using two or three end-member isotope mixing models to determine the distribution and sources of OM in estuaries, marginal seas, and continental shelves [e.g., (Cifuentes et al., 1996; Usui et al., 2006; Yu et al., 2015)]. However, conventional mixing models have critical limitations: 1) unable to solve the case where the number of sources exceeds the number of isotopes plus one; 2) unable to incorporate the variance of source isotopic composition; and 3) unreliable result with overlapping source isotopic composition (Davis et al., 2015; Phillips and Gregg, 2003). Mass balance mixing models such as IsoSource was developed to solve mixing of multiple sources but did not account uncertainty of source isotopic composition (Phillips and Gregg, 2003; Phillips et al., 2014). Recently, Bayesian models such as mixing models using sampling-importance-resampling (MixSIR) and Stable Isotope Analysis in R (SIAR) were developed to apportion the biogeochemical sources in mixed samples (Davis et al., 2015; Moore and Semmens, 2008; Phillips et al., 2014). Such model can apportion more than three sources when only two isotopes are used, and characterize the ranges and distributions of sources instead of using a single signature to represent one source. The most obvious advantage

* Corresponding author.
E-mail address: hzhang@yic.ac.cn (H. Zhang).

http://dx.doi.org/10.1016/j.marpolbul.2017.04.037
Received 26 January 2017; Received in revised form 17 April 2017; Accepted 21 April 2017
Available online 28 April 2017
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is that Bayesian can provide complete source proportional contributions, including the variance, mean value and quintiles of contributions. The Bayesian models were extensive applied to the investigation of trophic relationship in the food wed and recently been used in source apportionment of sediments (Cooper et al., 2015; Kubo and Kanda, 2017; Morris et al., 2015; Nosrati et al., 2014).

Thus the main objectives of this study are 1) to evaluate the relative contribution of riverine discharge and atmospheric deposition to sediment organic material in a semi-enclosed sea; and 2) to test the applicability of isotopic mixing model (MixSIR) for source apportionment of sediment organic material. The study is conducted in the Bohai Sea in northeast China, which is a semi-enclosed shallow sea with tremendous anthropogenic influences.

2. Materials and methods

2.1. Study area

Bohai Sea (37° 07′ ~ 41°0′ and 117° 35′ ~ 121° 10′) includes Laizhou Bay, Bohai Bay, Liaodong Bay, Bohai Strait and the Central Part. The shoreline of Bohai Sea has an overall length of 3780 km. The area of open water is about 7.7×10^4 km^2. Bohai Sea is a typical semi-enclosed shallow marginal sea of western Pacific Ocean, surrounded by several industrialized cities. The Bohai Strait with 80.5 km wide on its eastern side is the only passage connection to the Yellow Sea. The average depth is about 18 m and the deepest site (80 m) is located in the Laotieshan Channel of the Bohai Strait (Fig. 1). Bohai Sea has irregular semi-diurnal tide, subjected to strong tidal currents. It is influenced by monsoon climate, prevailing northwest wind in winter and southeast wind in summer. The averaged annual precipitation in the Bohai Sea is 500–600 mm, the overwhelming high precipitation occurs in summer, with an average value of ~400 mm. Terrestrial contributions to the sea are mainly through large rivers with total runoff about 2.0×10^{10} m^3/yr in year 2015. Among them, Yellow River contributes the highest runoff to the Bohai Sea with a discharge of 1.27×10^{10} m^3, accounts for about 60% of total discharge. During the Chinese economy boom in the last three decades, the circum-Bohai region has become a hub of industrialization and urbanization. The study area has been increasingly affected by air pollution from large-scale urbanization, industrialization and agriculture activities (Cao et al., 2006; He et al., 2015). Especially the North China Plain on the southwest of the Bohai Sea, which includes the two municipalities of Bejing and Tianjin and five provinces of Hebei, Henan, Shandong, Anhui and Jiangsu, have encountered the most severe air pollution in the last decade, due to heating in winter and straw burning (Fang et al., 2015; Zong et al., 2015). In addition, as the largest offshore crude oil production base in China, the Bohai Sea has numerous oil wells and drilling platforms under operation.

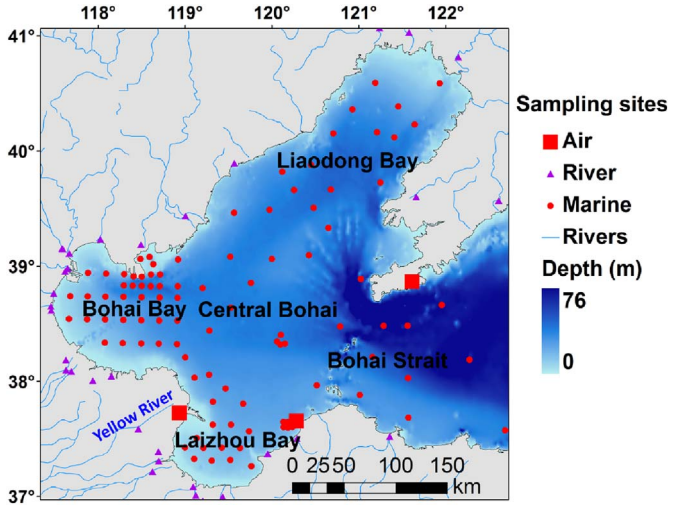

Fig. 1. Site locations of surface sediment samples (red dots), river particulate samples (purple triangles), atmospheric particulate samples (red squares) overlain on a bathymetric map of the Bohai Sea. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

2.2. Sample collection

Surface marine sediment (top 3 cm depth) samples were collected using a stainless steel box corer sampler during the summer cruise in August 2014 at 99 locations in the Bohai Sea. These sample sites were distributed in the nearshore and offshore zones, covered most parts of the sea (Fig. 1). At each site, water temperature, salinity, and chlorophyll *a* fluorescence at different water depth was measured in situ using CTD (SBE 911plus, USA). Measured salinity values of marine sampling sites were between 28 and 31 psu.

Sediment samples were collected in polyethylene bags, sealed and stored in a freezer until further analysis. Sediment samples for chemical analysis were freeze-dried, homogenized and ground to a fine powder with an agate pestle and mortar. The vegetation debris was removed before the grinding process. A portion of the homogenized fine powder samples was placed in glass tubes and acidized using 1 M hydrochloric acid to remove carbonate. After soaking for 12 h, hydrochloric acid was replaced for another 12 h until no air bubbles in tubes. The sample was then washed with deionized water to remove salts until becoming neutral and dried at 50 °C in a constant temperature oven.

Water samples in 36 rivers around the Bohai Sea were collected using Niskin bottles in May, August, and December 2015. Based on measured salinity values (< 1 psu), all river samples are freshwater devoid of sea water influence. Total suspended particulate matter (TSM) were filtered (0.3–1.5 L) on precombusted (450 °C for 24 h), constant temperature (25 °C) and humidity (39%) for 24 h, and preweighed Whatman GF/F filters and then stored at − 20 °C in refrigerator. After dried at 60 °C for 48 h, the samples were treated in the same condition of blank filters before being weighed on balance (Sartorius BSA224S, Germany) [e.g., (Sarma et al., 2014)]. A portion of dried samples were used for particulate nitrogen (PN) analysis. Filter samples for analysis of particulate organic carbon (POC) were dried at 50 °C for 24 h after removal of carbonated using HCl fumigation for 3 days (Hedges and Stern, 1984).

Sampling of total suspended particulates (TSPs) in the air was conducted in Dalian (the station in Dalian Maritime University), Dongying (the Yellow River Delta Ecological Research Station of Coastal Wetland), and Longkou (the Longkou Environmental Monitoring Station of the State Ocean Administration of China) during 2013–2015 as shown in Fig. 1. TSP samples were collected on separate quartz filters using a high volume air sampler (Tisch Environmental, Inc., USA) at a flow rate of 0.3 m^3/min.

2.3. Chemical and stable isotope analysis

The marine sediment, river TSM, and atmospheric TSP samples were analyzed for the total organic carbon (TOC) and total nitrogen (TN) via high temperature combustion on a CHNS analyzer (Elementar vario MACRO cube, Germany). Duplicate analyses of every ten samples were implemented. Replicate analysis gave a 1 σ precision of ± 0.4 wt % C and ± 1.3 wt% N. Standards used for carbon and nitrogen analysis include standard organic analytical standards (glutamate) and the national standard of soil (GBW 07434). The precision of this method based on replicate measurements (n = 6) of a reference standard is 0.8% for carbon and 1.3% for nitrogen.

Sediment samples powder and a portion of the carbonate free samples were analyzed by an isotope ratio mass spectrometer (Finnigan DELTA^{plus} XL, USA) for the measurement of the δ^{15}N and δ^{13}C values, respectively. The results are expressed as δ^{13}C and δ^{15}N in parts per thousand (permil; ‰), stable isotope ratios are reported in δ notation,

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