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# Source apportionment and source-to-sink transport of major and trace elements in coastal sediments: Combining positive matrix factorization and sediment trend analysis

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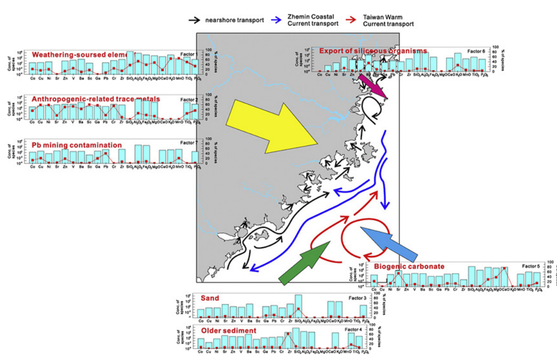
## HIGHLIGHTS

- Elemental concentrations and enrichment were assessed in coastal marine sediments.
- PMF was used for source apportionment and seven potential sources were identified.
- Net sediment transport patterns were established using the STA model.
- Patterns of elements' contribution coincide with source-to-sink transport processes.
- Transport of sediment as well as elements is interpreted in terms of hydrodynamics.

## GRAPHICAL ABSTRACT

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## ABSTRACT

Major and trace elemental concentrations in coastal marine sediments were incorporated into positive matrix factorization (PMF) to identify potential sources and source contributions. Transport pathways of fine-grained sediments and sediment-bound elements were inferred from sediment trend analysis (STA). The spatial distribution patterns of 21 elements (Co, Cu, Ni, Sr, Zn, V, Ba, Sc, Ga, Pb, Cr, Zr, $SiO_2$, $Al_2O_3$, $Fe_2O_3$, MgO, CaO, $K_2O$, MnO, $TiO_2$, and $P_2O_5$) coupled with sediment grain sizes were investigated. The natural and anthropogenic sources of the elements were distinguished by their medium enrichment factors (EFs). Seven sources were recognized by PMF: weathering products, anthropologic emissions, sand, older sediment, biogenic carbonates, products of siliceous organisms, and mine exploitation. Some land-derived elements, including weathering products, anthropogenic-related elements, and mining-related elements, had a significant positive correlation with sediment silt, clay, and organic carbon contents. The spatial patterns of the land-derived elements' concentrations and source contributions were consistent with the sediment transport pathways inferred from the STA. This result revealed that the delivery of the land-derived elements was determined by marine current flows and the associated sediment transport processes. Conversely, elements originating from marine sources, such as sand and older sediment, and from the biological activities of calcareous and siliceous organisms showed little response to sediment transport and deposition processes. Our study links the outputs of statistically oriented approaches (e.g., PMF) to a process-based understanding of elemental transport in marine environments.



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## 1. Introduction

Trace metal contamination in estuaries and coastal areas has recently received increasing attention because it may harm humans, living resources, and marine ecosystems. Metals accumulate in sediments and can be absorbed by benthic organisms or released back into the water, causing water quality degradation (Pan and Wang, 2012). The sources of trace metal contaminants in coastal sediments are mostly based on land, while a small fraction of these contaminants originate from marine-based sources (Weis, 2014). Understanding the sources of trace metals and the related characteristics may provide useful information for policy makers to effectively manage coastal resources and habitats.

Multivariate statistical approaches, such as cluster analysis (CA), principal component analysis (PCA), and PCA-related methods, are usually used to infer potential sources from geochemical properties of sediments (Jiang et al., 2017; Owens et al., 2016). Factor loadings and factor score matrices obtained from PCA are interpreted as source composition and source contributions, respectively. These loadings and scores can be positive or negative. Since negative compositions and negative contributions do not actually exist, it is difficult to quantitatively assess the relative contributions from the sources identified.

An alternative approach is imposing nonnegativity constraints on the resulting factors to avoid the effect of the negative values (Paterson et al., 1999). Positive matrix factorization (PMF) is an advanced factor analytical method that fits the nonnegativity criterion (Paatero and Tapper, 1994; Vaccaro et al., 2007). PMF can efficiently handle measurement uncertainties, outliers, values below measurement detection limits, and missing values (Hemann et al., 2009), thus offering a favorable method for environmental applications. Individual data points are weighted so that both small and large data are modeled correctly, and in particular, outliers do not overly influence the forming factors (Bzdusek and Christensen, 2006; Bzdusek et al., 2006). By contrast, outliers are treated as noisy data or non-representative data and are sometimes omitted from PCA. In addition, PMF factors do not need to be orthogonal, which is important for linking the modeled factors to the real sources (Hemann et al., 2009). Numerous studies have quantitatively inferred the sources of trace metals within a wide range of materials, such as atmospheric sediment (i.e., dust and airborne particulate matter), soil, and river deposition (Comero et al., 2012; Comero et al., 2014; Hsu et al., 2016; Jiang et al., 2015; Mazzei et al., 2008; Praẑnikar et al., 2014; Tecer et al., 2012; Zhang et al., 2018).

Once trace elements are released into an aquatic system, they adhere to suspended fine-grained sediment particles (Salomons and Stigliani, 1995) or incorporate into the organic matter fraction (Hirner et al., 1990). Suspended particles are entrained in currents and transported in the same direction as the currents. To obtain further insight into the transport properties of sediments and sediment-bound trace metals, sediment trend analysis (STA) is performed. This method attempts to infer the net sediment transport pattern using grain size trends (McLaren, 1981; McLaren et al., 2007). In most cases, STA reflects the movement of fine-grained sediment in a hydrological system since finer (less dense) grains are preferentially mobilized and transported. Our previous works (Li et al., 2015a; Li et al., 2015b; Li et al., 2013; Li et al., 2014) have demonstrated that there is a direct connection between high trace metal concentrations (e.g., Ba, Co, Cr, Cu, Ni, Pb, V, and Zn) and high percentages of fine-grained sediments, particularly those in silt and clay fractions. Notable decreases in the concentrations of sediment-associated metals occur along the sediment transport pathways. This finding suggests that sediment transport processes have implications for trace metal delivery.

The middle and southern Fujian coastal area, located on the west side of the Taiwan Strait, is an important farming area in Fujian Province. This area faces increasing metal pollution problems due to the rapid industrialization and urbanization that has occurred over the past two decades. Metal pollution may threaten seafood safety, and it was reported that cultured-shellfish products from this area are contaminated with heavy metals (Xi et al., 2017). Many studies have been conducted on the concentrations, distribution, and ecological risk of trace metals from surface sediments in a number of bays (and estuaries) along the middle and southern Fujian coast (Cai, 2010; Cai et al., 2007; Chen, 2008; Chen et al., 2014; Huo et al., 2015; Li et al., 2008; Li et al., 2010; Lin, 2008; Lin, 2012). In these studies, trace metals were assumed to originate from rock weathering and agricultural, mining, and industrial processes. Nevertheless, the compositions and quantitative contributions of the potential sources are not well documented.

Although much research has been invested in identifying and quantifying sources of trace metal contamination, only a few studies have been conducted on estuarine and coastal sediments. In this study, we performed PMF to (1) distinguish the different sources of 21 trace and major elements in the middle and southern Fujian coastal sediments and (2) quantitatively evaluate the composition and relative contributions of each source. Some of the 21 elements, e.g. toxic metals, may be enriched in seafood and enter the human body through the food chain, posing a great threat to human health. Others compounds, such as land-derived nutrients ($P_2O_5$), can potentially cause eutrophication and adverse ecological effects. To specifically understand how sediment properties change during transport processes, changes in source contributions were linked to the net sediment transport pattern derived from STA. To our knowledge, this combination of PMF and STA has not been used for the interpretation of the transport dynamics of metals in aquatic settings, and we believe that our approach has valuable applications.

## 2. Study area

The study area is situated on the western part of the Taiwan Strait (Fig. 1). The coastline morphology is oriented NW–SE, with the northern section facing N–W. The topography of the sea bottom coincides with the coastline, and the isobaths are generally parallel to the coast (Fig. 1). The geographic and hydrodynamic characteristics of this area have been documented elsewhere (Li et al., 2018). Here, we highlight the river hydrology in the adjacent hinterlands, and most of our data are collected from the Annals of Fujian Gulfs (The Annals of Chinese Gulfs Compiling Committee, 1993, 1994).

Eight rivers and streams flow into the coastal bays and estuaries, including Saijiang, Huotongxi, Qibuxi, Aojiang, Minjiang, Mulanxi, Jinjiang, and Jiulongjiang (Fig. 1). The Minjiang River is the largest river of Fujian Province, with an average annual discharge of 58,600 $Mm^3$ and an annual suspended sediment discharge of 7.155 Mt. The Jiulongjiang River is the second largest river of Fujian Province, and it has a drainage basin of 14,741 $km^2$, an average annual discharge of 12,110 $Mm^3$, and an annual suspended sediment discharge of 2.23 Mt. The other rivers and streams have a small drainage area. The drainage basin areas of Jinjiang, Saijiang, Aojiang, Mulanxi, and Qibuxi are 5692 $km^2$, 5549 $km^2$, 2655 $km^2$, 1732 $km^2$ and 222.48 $km^2$, and their average annual discharges are 5502 $Mm^3$, 4140 $Mm^3$, 3040 $Mm^3$, 985 $Mm^3$, and 134 $Mm^3$, respectively.

Bathymetrically, the study area can be divided into four parts: (1) a nearshore zone at a water depth <20 m, (2) two estuaries, (3) a coastal zone where the water depth varies between 20 m and 40 m, and (4) a continental shelf in the southeastern part of the study area (deeper than 40 m).

## 3. Methods

### 3.1. Sampling and measurements

Surface sediment samples were collected at 1172 sites during four separate marine surveys from 2012 to 2016, and the water depths