Estuarine, Coastal and Shelf Science 121-122 (2013) 61–68

Contents lists available at SciVerse ScienceDirect

Estuarine, Coastal and Shelf Science

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

# Determination of water ages and flushing rates using short-lived radium isotopes in large estuarine system, the Yangtze River Estuary, China

Bo-Chao Xu [a,b], Natasha T. Dimova [c], Liang Zhao [d], Xue-Yan Jiang [a], Zhi-Gang Yu [a,*]

[a] Key Laboratory of Marine Chemistry Theory and Technology, Ministry of Education, Ocean University of China, Qingdao 266100, China
[b] Key Laboratory of Marine Environment and Ecology, Ministry of Education, Ocean University of China, Qingdao 266100, China
[c] Department of Geological Sciences, University of Alabama, Tuscaloosa, AL 35406, USA
[d] Physical Oceanography Laboratory, Ocean University of China, Qingdao 266100, China

ARTICLE INFO

*Article history:*
Received 11 October 2012
Accepted 11 February 2013
Available online 26 February 2013

*Keywords:*
Yangtze River Estuary (YRE)
radium isotopes
water ages
water circulation
river basin management
modeling

ABSTRACT

We investigated the spatial and temporal distribution of naturally-occurring short-lived radium isotopes ($^{224}$Ra, $t_{1/2} = 3.6$ d and $^{223}$Ra, $t_{1/2} = 11$ d) to examine coastal water mixing dynamics of the third world largest estuary, Yangtze River Estuary (YRE) during two field trips in April 2010 and May 2011. Distributions of the $^{224}$Ra/$^{223}$Ra activity ratios within the YRE area were used to calculate apparent estuarine water ages. Field-derived results were then compared to hydrodynamic assessments obtained by a Lagrangian particle tracking simulation experiment performed using the Princeton Ocean Model (POM). Water ages obtained via both geotracers and particle tracking agree very well. During both field trips an anomalously "younger" water mass (low salinity and higher radium activities) was observed at about 90−170 km offshore distance from the mouth of the river, suggesting an additional terrestrial water source influenced this area. The temporal distribution of the radium isotopes indicated a semi-diurnal tidal pattern in the YRE with relatively constant isotopic composition of less than a 20% variation during our observations. An integrated water flushing rate based on our observations (excluding the additional anomalous source area) was 8.4 km day$^{-1}$.



## 1. Introduction

Estuaries are important transition zones between land and ocean. Both fresh water and recirculating seawater are found to provide high levels of nutrients to the water column, making estuaries among the most productive natural habitats in the world (Houde and Rutherford, 1993; Harding et al., 2002; Burford et al., 2008; Sukhanova et al., 2010). Water residence times and salinity variations directly influence the ecological conditions and production rates in estuaries (Dulaiova and Burnett, 2008). Thus, estimates of coastal water residence times are extremely important when studying chemical and biological changes occurring within these areas and in evaluating offshore fluxes of dissolved terrestrial materials that reach the open ocean (Moore, 2000). Small-scale temporal and spatial variability due to precipitation, evaporation, as well as water management operations contribute to the high level of complexity in the estuarine environments. Developing an understanding of the physical and biogeochemical processes in these systems is a challenge for coastal oceanographers and water managers (Moore and Krest, 2004).

Short-lived radium isotopes, $^{224}$Ra ($t_{1/2} = 3.66$ days) and $^{223}$Ra ($t_{1/2} = 11.4$ days), have become increasingly popular for examining mixing processes in coastal zones over time scales of days to weeks (e.g., Moore, 2000, 2006; Peterson et al., 2008a; Knee et al., 2011). In fresh water radium is fairly particle active ($K_d \approx 10^3$) resulting in very low concentrations (Raanan et al., 2009). However, in brackish and salt water the adsorption coefficient of radium decreases rapidly ($K_d \approx 10^1$) as the water ionic strength increases (Langmuir and Riese, 1985; Webster et al., 1995; Rama and Moore, 1996; Gonneea et al., 2008) resulting in much higher radium concentrations in coastal areas. Desorption of radium from particles occurring within minutes of exposure to saline high ionic strength water makes up to 30−50% of the total desorbed radium (Gonneea et al., 2008; Kiro et al., 2012). Other source of radium could include submarine groundwater discharge (SGD) although this is likely to have a relatively minor contribution in river-dominated estuaries (Krest et al., 1999; Moore and Krest, 2004; Dulaiova and Burnett, 2008; Peterson et al., 2008a). In general, the distribution of short-lived radium in the surface waters of an estuary is governed by

* Corresponding author. Key Laboratory of Marine Chemistry Theory and Technology, Ministry of Education, Ocean University of China, Qingdao 266100, China.
*E-mail address:* zhigangyu@ouc.edu.cn (Z.-G. Yu).

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http://dx.doi.org/10.1016/j.ecss.2013.02.005

mixing and radioactive decay: as water flows offshore driven by river plume and near-shore oceanic currents, it becomes depleted in radium due to mixing with radium depleted ocean water as well as absence of parent to support radium production in the water column. Therefore, in principle, one could predict trajectories of oceanic currents within coastal zones by examining radium presence in these coastal waters.

With a total length of 6300 km, the Yangtze River (Changjiang) is the third longest river in the world. It drains a land area of $1.8 \times 10^6$ km$^2$, which is about one fifth of the total land area of China. Historically (between the 1950s and 2005), the river used to deliver $9.0 \times 10^{11}$ m$^3$ of runoff discharge and $4 \times 10^8$ tons of sediment load per year to its estuary and the East China Sea (ECS, Wang et al., 2008a). It is predicted that the continued extensive program of dam constructions within the Yangtze River catchment, including the vast Three Gorges Dam, will continue to reduce the quantity of material exported by the river (Tim et al., 2010). The Three Gorges Dam's construction was completed in 2006 and its designed maximum reservoir water level of 175 m was first reached on 26 October, 2010. In the future, the water level will be kept between 145 m and 175 m, depending on flood control needs. The inter-tidal wetlands of the Yangtze River delta are listed as one of the world's most important wetland ecosystems and have been designated as a Chinese Natural Reserve (Yang et al., 2005). The Zhoushan fishing ground, the fourth largest fishery base in China, is also in the zone influenced by the discharge of the river (Yang et al., 2002). Changes to the water circulation within the continental shelf and in the coastal zone as a result of changing water residence times caused by reduced river discharges may have large scale ecological and economical impacts.

The YRE and adjacent ECS represent a very complex hydrological system, which is perennially governed by some major current systems (Fig. 1): the Yellow Sea Coastal Current (YSCC), the Yellow Sea Warm Current (YSWC), the Taiwan Warm Current (TWC) and the Zhejiang-Fujian Coastal Current (ZFCC). The dynamics of the system has been mainly evaluated by numerical modeling under certain assumptions (Ichikawa and Beardsley, 2002; Lü et al., 2006; Chen et al., 2007). Isotopic tracing, on the other hand, provides estimates based on real-time field measurements and thus may provide more realistic evaluations of the water dynamics in the YRE.

The main purpose of this study was to examine the water circulation within the inner continental shelf of the YRE using spatial and temporal distribution patterns of $^{223}$Ra and $^{224}$Ra and their ratios. The estuarine "water ages" were evaluated by the "apparent radium age" model based on spatial and temporal distributions of $^{224}$Ra to $^{223}$Ra activity ratios (Moore, 2000). For comparison, a Lagrangian particle-tracking simulation experiment driven by the Princeton Ocean Model (POM) was also used to track the fate of the Yangtze River diluted water mass. This work will demonstrate that the "water age" model can be applied to large estuaries with complex hydrodynamics such as the YRE. Furthermore, the information obtained here could be applied to help developing management strategies for controlling the supply of terrestrial nutrients and contaminants to offshore areas and protect the environment of this area.

## 2. Material and methods

### 2.1. Sampling and analytical methods

Samples were collected during two expeditions in April 2010 and May 2011 (see sampling stations shown in Fig. 1). Because of restricted access to the YRE and the river in April 2010, water samples from the river mouth (i.e., river end-member) were not collected. As a result, only nine transect samples with salinities between 20 and 31 were collected. In order to investigate the temporal distribution of radium, water was also collected in two time-series (T1 and T2) of 15 and 24 h, respectively. During the second field trip in May, 2011 full access to the upstream YRE area allowed us to collect more water samples in a full range of salinities (e.g., 0–31). The average river discharges during the sampling intervals were very similar, $19\ 800 \pm 700$ m$^3$ s$^{-1}$ and $18\ 720 \pm 311$ m$^3$ s$^{-1}$ respectively, while historical annual discharges averaged 28 935 m$^3$ s$^{-1}$ for the period between the 1950s and 2005 (Wang et al., 2008a).

A routine radium sampling procedure was applied during both trips: large volume surface water samples (~200 L) were collected

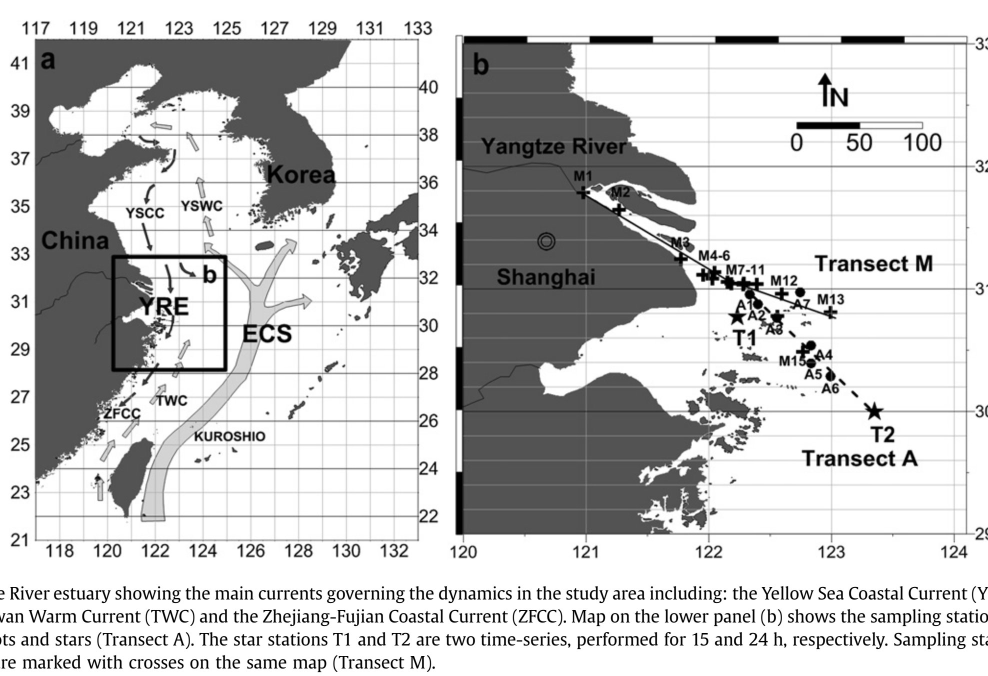


**Fig. 1.** Map (a) of Yangtze River estuary showing the main currents governing the dynamics in the study area including: the Yellow Sea Coastal Current (YSCC), the Yellow Sea Warm Current (YSWC), the Taiwan Warm Current (TWC) and the Zhejiang-Fujian Coastal Current (ZFCC). Map on the lower panel (b) shows the sampling stations during the field work in April 2010 marked by dots and stars (Transect A). The star stations T1 and T2 are two time-series, performed for 15 and 24 h, respectively. Sampling stations during the sampling campaign in May 2011 are marked with crosses on the same map (Transect M).