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Radium isotopes and their environmental implications in the Changjiang River system

Ni Su [a, b], Jinzhou Du [a, *], Zonglian Duan [a], Bing Deng [a], Jing Zhang [a]

[a] State Key Lab of Estuarine and Coastal Research, East China Normal University, Shanghai 200062, PR China
[b] State Key Lab of Marine Geology, Tongji University, Shanghai 200092, PR China

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A B S T R A C T

Radium (Ra) isotopes are widely used to trace water motion and nutrient transport in coastal oceans. To enhance our understanding of Ra behavior from river to ocean, this study examines ^{226}Ra and ^{228}Ra activity concentrations in the water column, suspended particulate matter and river bed surface sediments along the middle and lower Changjiang River (CJR) basin based on two cruises in 2006 and 2008. The results show that the Ra activity concentrations in the CJR basin are comparable with other rivers in the world. Higher Ra activity concentrations and lower ^{228}Ra/^{226}Ra activity ratios are detected in the tributaries (i.e., in the Dongting and Poyang Lakes) than in the mainstream. The comparison of pre-Three Gorges Dam (TGD) data (taken in 1984) and post-TGD data (this work) illuminates the effect that a major dam has on radium transport within the river and into the estuary. We estimate a riverine ^{226}Ra flux of 1.04 ± 0.08 TBq yr^{-1}. Extended Ra datasets collected in the estuary and the East China Sea show that the dissolved and sedimentary Ra activity concentrations are higher in the estuary but lower in the sea, whereas the particulate Ra activity concentrations are larger in the river. This finding indicates significant Ra desorption from particulate to dissolved phase in the brackish estuary where riverborne materials are filtered and trapped. Overall, these results help to elucidate Ra transport from river to ocean and assist further application of Ra isotopes in land–ocean interaction studies, such as submarine groundwater discharge, mixing and diffusion processes, and determination of water mass and/or current structures in the estuarine and coastal zones.

1. Introduction

Riverborne particulate and dissolved material, e.g., nutrients and some radioactive nuclides, are of significant interest in land–ocean interaction studies. There are already numerous reports on the riverine fluxes of, for instance, sediments, nitrogen and silica, whereas less scientific attention has been devoted to radionuclides (Shen, 2001; Beusen et al., 2005; Syvitski et al., 2005). Radioactive isotopes are used as tracers in examining coastal ocean processes. In this study, we surveyed radium (Ra) activity concentrations in the Changjiang River (CJR) basin of China to explore its environmental implications, such as the variations in activity concentrations and distribution of Ra isotopes in the CJR basin due to the environmental change (i.e., the construction of dams). The interest in Ra lies in the half-lives of the isotopes that stretch from days to thousands of years, thereby enabling their application over a wide range of timescales. Also, Ra isotopes are increasingly used in tracing coastal oceanography processes, especially submarine groundwater discharge (Moore and Scott, 1986; Nozaki et al., 1991; Moore, 1996, 2000).

Ra isotopes, including ^{223}Ra ($T_{1/2} = 11.4$ days), ^{224}Ra ($T_{1/2} = 3.66$ days), ^{226}Ra ($T_{1/2} = 1622$ years) and ^{228}Ra ($T_{1/2} = 5.75$ years), are members of the uranium and thorium series. Uranium and thorium are present in large quantities in the Earth's crust (Osmond and Ivanovich, 1992). Ra isotopes in particular ^{223}Ra, ^{224}Ra and ^{228}Ra are present at very low abundance only, due to their short half-lives. As a result of the long-term weathering of rocks, Ra isotopes are released and transported via water and sediment in rivers before ending up in the oceans. Ra isotopes also exhibit particle reactive properties, meaning they are strongly adsorbed onto riverine particles in freshwater, but can be desorbed from suspended particles in a brackish environment (i.e., an estuarine mixing zone) with increasing salinity, known as non-conservative behavior (Elsinger and Moore, 1980; Key et al., 1985). These

* Corresponding author.
E-mail address: jzdu@sklec.ecnu.edu.cn (J. Du).

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properties can provide useful insights into associated geochemical processes. There are reports on Ra activity concentrations in rivers such as the Mississippi, the Amazon and the Ganges-Brahmaputra (Moore and Edmond, 1984; Moore and Scott, 1986; Sarin et al., 1990; Krest et al., 1999; Moore and Krest, 2004), but there is little data concerning other large rivers with significant geological and environmental features. In the case of China, there have been researches performed concerning the application of Ra in the CJR estuary and adjacent sea (Elsinger and Moore, 1984; Zhang, 2007; Peterson et al., 2008; Men et al., 2010; Xu, 2011; Gu et al., 2012; Su et al., 2013). Measurements of ^{226}Ra were reported in a basic survey of radioactivities in 1984 in the CJR basin (Li, 1984). Since then, there has been no systematic data showing the abundance of Ra in the CJR basin and its impact after the building of the Three Gorges Dam (TGD), especially its transport from the river to the sea.

In this study, we collected samples in the water column and surface sediments during two cruises along the CJR mainstream and major tributaries in the middle and lower CJR basin, aiming to understand the background activity concentrations of Ra and how the TGD has affected radium transport within the watershed. We also examined the literature concerning Ra data in the CJR estuary (CJE) and the East China Sea (ECS) to determine the transport behavior of Ra from river to ocean with respect to land–ocean interactions.

2. Field work and methods

2.1. Study area

The CJR is the longest river in China with a length of about 6300 km and a drainage area of 1.8 million km^2. Geographically, the CJR basin is divided into an upper basin stretching from the river head to Yichang, a middle basin between Yichang and Jiujiang, and a lower basin between Jiujiang and Datong (see Fig. 1). The upper CJR basin is occupied by high relief mountains (with elevations greater than 5000 m) and incised valleys, and the middle and lower CJR basins mainly contain low relief fluvial flood plains. Downstream of Datong, the CJR enters an estuary before finally emptying into the ECS.

The CJR has a huge runoff and sediment load, annually 903.4 km^3 and 414 Mt at Datong, respectively (1950–2005) (Chen et al., 2008). The annual mean precipitation is about 1100 mm over the whole basin (Yang et al., 2007a). Seasonally, approximately 70% of the cumulative flow and 85% of the sediment load are discharged during the wet season between May and October (Chen et al., 2008). Spatially, half of the river discharge at Datong originates from the upper CJR basin and the remaining part is from the tributaries in the middle and lower basins (i.e., the Dongting Lake, the Poyang Lake and the Hanjiang River). The upstream basin also

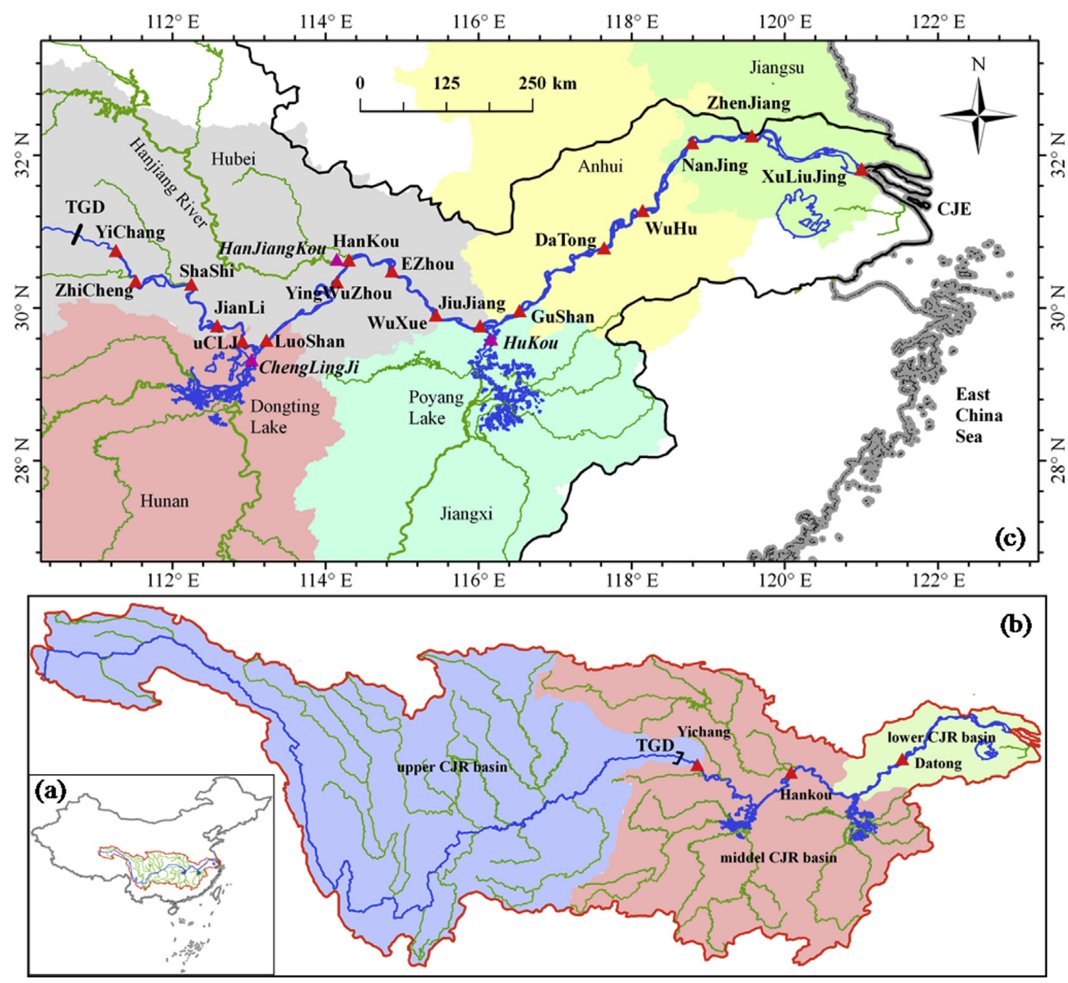

Fig. 1. (a) Outline of mainland China; (b) the Changjiang River basin; (c) sampling stations in the middle and lower reaches of the Chanjiang River basin in October 2006 and January 2008. TGD means the Three Gorges Dam. The italic names refer to the stations in tributaries.