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Distribution of radionuclides in a marine sediment core off the waterspout of the nuclear power plants in Daya Bay, northeastern South China Sea

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ABSTRACT

A sediment core was collected and dated using $^{210}Pb_{ex}$ dating method off the waterspout of nuclear power base of Daya Bay, northeastern South China Sea. The γ-emitting radionuclides were analyzed using HPGe γ spectrometry, gross alpha and beta radioactivity as well as other geochemical indicators were **deliberated** to assess the impact of nuclear power plants (NPP) operation and to study the past environment changes. It suggested that NPP provided no new radioactivity source to sediment based on the low specific activity of ^{137}Cs. Two broad peaks of TOC, TC and LOI accorded well with the commercial operations of Daya Bay NPP (1994.2 and 1994.5) and LNPP Phase I (2002.5 and 2003.3), implying that the mass input of cooling water from NPP may result into a substantial change in the ecological environment and Daya Bay has been severely impacted by human activities.

1. Introduction

Marine sediment serves as one of the major reservoirs for all kinds of contaminants, and it is a useful natural archive containing a wealth of environmental information and providing a high-resolution chronology of pale-environmental proxies. Sediment preserves a useful natural archive that enables the study of the environment changes and anthropogenic disturbances from the past. Thus the dated sediment samples allow studies of past natural and anthropogenic environmental conditions and changes (Morgenstern et al., 2001; Vongunten et al., 1997). $^{210}Pb_{ex}$ ($T_{1/2} = 22.3$ year) dating method has been applied to the measurement of the sedimentation rates in lakes, estuaries, and coastal marine sediments (Goldberg, 1963; Goldberg et al., 1977; Robbins and Edgington, 1975; Huang et al., 1983; Demaster, 1985; Murchie, 1985; Chen et al., 1990; Ivanovich and Harmon, 1992; Sanchez-Cabeza and Ruiz-Fernández, 2012), which is the most suitable for determining the chronology of the past 100 years in sediment cores to establish the timing of the past environmental changes and to assess the accumulation of materials (Li, 1993; Fuller et al., 1999; Ruiz-Fernández et al., 2002; Ribeiro Guevara et al., 2003; Kato et al., 2003; Ayçik et al., 2004; Li et al., 2006; Dai et al., 2007; Yang et al., 2009; Ruiz-Fernández and Hillaire-Marcel, 2009; Vrel et al., 2013).

As one of the biggest inlets and an important cultural area in Guangdong province, China, Daya Bay is a subtropical drowned valley bay in the north-east part of Pearl River estuary, which locates in the north-east part of the South China Sea. Daya Bay is a semi-enclosed shallow embayment and is separated into several small sub-basins, with surface area of 650 km^2 and a coastal line of 92 km. Its water depth ranges from 6 to 15 m with the average water depth of 10 m (Xu, 1989; Wang et al., 1996; Du et al., 2008). The climate of Daya Bay gives mild, wet subtropic weather with the annual variations of water temperature from 14.0 °C (winter) to 32.8 °C(summer) and an average of 23.5 °C. Salinity is usually between 22.7 and 33.8, and fluctuates little, except in the typhoon

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season. Its current is dominated by tides and surface water has an average resident time of about 3.2 days. Daya Bay has been one of the main aquaculture areas in Guangdong Province thanks to its rich biological due to its excellent water quality resources (Wang et al., 2004a, 2006a).

However, Daya Bay has been severely impacted by human activities during the past more than 30 years. Petrochemical, plastic, printing and other industries, as well as harbors and aquaculture are present around the sea area. Two nuclear power plants (also named DNPP base), Daya Bay Nuclear Power Plant (DNPP) (2 × 984 MW) and Ling'ao Phase I Nuclear Power Plant (LNPP Phase I) (2 × 990 MW), which came into operation in 1994 and 2002 respectively, are situated at its western coast. The warm water from DNPP base main effect the surface sea water temperature, the increasing temperature of sea water maybe influence on the carbon cycle of the region directly (Wang et al., 2004a). Massive economic growth and urban development in the region have led to excessive release of waste into the bay. They may be causing water quality deterioration. However, no overall study has been conducted on the environmental effects caused by natural and anthropogenic activities in the bay (Peng et al., 2002; Wang et al., 2004a, 2004b; Song et al., 2004; Liu et al., 2006; Wang et al., 2006a, 2008a, 2008b; Wu and Wang, 2007; Zhou et al., 2001; Gao et al., 2010).

In this paper, in order to reconstruct the long-term history of the environment changes, and to evaluate whether the nuclear power plants have some radioactive or non-radioactive effect to the environment in Daya Bay, a core was sampled off the waterspout of the nuclear power plants in Daya Bay. The specific activities of ^{238}U, ^{226}Ra, ^{210}Pb, ^{228}Th, ^{228}Ra, ^{40}K, ^{137}Cs in the sediment core were determined using HGe γ spectrometry and the mass accumulation rate was calculated by $^{210}Pb_{ex}$ dating method of constant flux constant sedimentation model (CF or CFCS) (Oldfield et al., 1978; Appleby et al., 1979; McCall et al., 1984). In addition, TOC, TC, LOI, gross alpha and beta radioactivity in this core were analyzed to understand this influence on marine environment from the DNPP base in this sea area.

2. Materials and methods

2.1. Description of sampling

The sampling station of the sediment core W2 (2) in the present work is shown in Fig. 1. A sediment core was collected using a gravity core of 110 mm inner diameter in Daya Bay on July 28th, 2010. As shown in Fig. 1, the sampling station W2 (2) (22°36. 20′N, 114°34.00′E) was located off the waterspout of GNPP and LNPP. The water depth was about 10.0 m. After the sediment core was sampled *in situ*, and then was transferred to the laboratory (GB 17378.3-2007, China) and immediately stored at −20 °C until analyzed. In the laboratory, the core was sub-sampled at 2.0 cm interval using a stainless steel blade for the isotopic and chemical analysis. The W2 (2) core with 60 cm length was sliced into 30 samples.

2.2. Analytical methods

2.2.1. Sediment granulometry

The sample granulometry was analyzed using a Malvern Mastersizer 2000 laser diffractometer capable of analyzing particle sizes between 0.02 μm and 2000 μm. The main technical requirements for the sediment grain size analysis were referred the standard of specifications for oceanographic survey (GB/T 12763.8-2007, China). The types of grain group were determined: clay (mud) (Y) (<0.001 mm–0.004 mm), silt (T) (0.004 mm–0.063 mm), sand (S) (0.063 mm–2 mm), gravel (G) (2 mm–256 mm), rock (R) (>256 mm).

2.2.2. Water content, loss-on-ignition (LOI)

All data were corrected for the sample's dry weight. The water content of the sediment was determined gravimetrically by comparing the weight difference before and after heating an aliquot at 105 °C until constant weight. The percentage of water was used to convert substance content of the sediment from wet to dry weight base (Li et al., 2007a). The loss-on-ignition (LOI) was estimated to the contents of total organic matter (TOM) in the sediment. After the water content was measured, the sample was heated at 550 °C until constant weight to estimate TOM (as a percentage of dry weight).

2.2.3. Total Organic Carbon (TOC), Total Inorganic Carbon (TIC), Total Carbon (TC)

Total Organic Carbon (TOC), Total Inorganic Carbon (TIC) and Total Carbon (TC) were determined using burning oxidation-non-dispersive infrared absorption method, carried out with a Shimadzu TOC-VCPH total organic carbon analyzer (SHIMADZU, Japan). The reference materials (RM) to determine TOC and TIC in sediment samples were by using glucose ($C_6H_{12}O_6$) (Guaranteed reagent) and Sodium bicarbonate ($NaHCO_3$) (Guaranteed reagent) (National Institute of Metrology P.R. China). Before the reference materials (RM) of glucose and Sodium bicarbonate being used, they should be dried at 105 °C until constant weight.

2.2.4. Radionuclides' determination

The measurement of γ-emitting radionuclides was performed at Radioactivity Monitoring Laboratory of *South China Sea Environment Monitoring Center, State Oceanic Administration* (SEMC, SOA). After the gravel and larger particles of biological debris were removed, the water contents were determined through drying by heat about 2.0 g of sample at 105 °C to constant weight for evaluating the dry weight of sample. The wet samples were sealed in air-tight plastic bag, then was placed into a plastic container with a height of 5.0 cm and diameter of 7.5 cm and pressed into cylindrical form with same diameter with the container. Before counting by gamma-spectrometry, the samples in air-tight plastic bag were kept for a period of 20 days to assure ^{226}Ra in the samples to reach secular equilibrium with its daughters ^{222}Rn (Liu et al., 2001; Chen et al., 2005; Li et al., 2007a; HY/T 003·8-91).

The measurements were performed using a HPGe γ-spectrometer with CANBERRA Model 747 Lead Shield and Model BE5030 (Broad Energy Ge) detector with its crystal diameter of 80.5 mm and thickness 31.0 mm (from Canberra USA). The integrated background count of system of Model 747 Lead Shield is 1.85 s^{-1} in the energy range of 10–1635 keV. The energy range of BE5030 detector from 3 keV to 3 MeV combines the spectral advantages of Low Energy and Coaxial HPGe detectors. The resolution at low energies is equivalent to that of Low Energy Ge (LEGe) Detector and the resolution at high energy is comparable to that of good quality coaxial (SEGe) detectors, so that the interferences in the low energy window was negligible when resolving the spectra (e.g., to determine ^{210}Pb and ^{234}Th). Resolution (FWHM- Full Width Ha lf Max) at 5.9 keV (^{55}Fe), at 122 keV(^{57}Co), at 1332 keV (^{60}Co), are 0.500, 0.750, 2.200 keV, respectively. The relative efficiency is about 50% for 1332 keV γ-ray of a ^{60}Co point source located 25 cm distance. The multi-channel analyzer is Accuspec interface board coupling with microcomputer and conversion gain and memory are 8192. Genie-2000 software was used to analyze spectrum data.

The ^{210}Pb activities were determined by the direct measurement of 46.5 keV gamma peak. The ^{40}K and ^{137}Cs radionuclides were measured from their respective γ-ray energies 1460 and 661.66 keV, respectively. The γ-ray energies of ^{208}Tl (583.2 keV) and ^{228}Ac (338.4, 911 and 969 keV) were used to determine the concentration of ^{228}Th and ^{228}Ra. The γ-ray energies of ^{234}Th (92.37