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Distribution of uranium isotopes in the main channel of Yellow river (Huanghe), China

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ABSTRACT

Uranium isotopes were measured in waters and suspended particulate matters (SPM) of the main channel of Yellow River, China that were sampled during four field trips between August 2005 and July 2006. The results show that the concentration of dissolved U (2.04–7.83 μg/l) and the activity ratio of $^{234}U/^{238}U$ (1.36–1.67) are much higher than the average U concentrations and activity ratios of global major rivers. Mass balance calculations using the results of simulated experiments and measurement data show that the section of the Yellow River between Lanzhou and Sanmenxia has its dissolved U derived from two sources: suspended sediments (68%) and groundwater/runoff from loess deposits (32%). Both sources are related to the heavy erosion of the Chinese Loess Plateau.

1. Introduction

Uranium-series isotopes serve as important research tools in geochronological, paleoenvironmental and geohydrological studies (e.g., Ivanovich and Harmon, 1992; Windom et al., 2000; Kronfeld et al., 2004). As a minor constituent of seawater, U has a remarkably uniform oceanic distribution (Sarin and Church, 1994), showing an average concentration (for seawater of salinity 35) of 3.3 ± 0.2 μg/l and a $^{234}U/^{238}U$ activity ratio of 1.144 ± 0.004 (Ku et al., 1977; Chen et al., 1986). Three main transport pathways supply U to the ocean: river runoff, direct groundwater discharge and aeolian dust with river runoff being the dominant supply (Palmer and Edmond, 1993; Chabaux et al., 2001; Dunk et al., 2002). The concentration of U varies widely in river waters, typically ranging over more than two orders of magnitude from 0.02 to >10 μg/l (Scott, 1982). Palmer and Edmond (1993) determined dissolved U in forty major rivers from around the world and obtained an average concentration of 0.31 μg/kg. They noted that this value could be biased by the very high levels observed in the Ganges–Brahmaputra and the Yellow River. Excluding these two river systems reduces the global average to 0.19 μg/kg.

Disequilibrium between ^{238}U and ^{234}U in natural waters and sediments has been found to be a rule rather than an exception. Mechanisms causing such disequilibrium include the α-particle recoil ejection of ^{234}Th (a precursor of ^{234}U) into solution (Kigoshi, 1971); the preferential chemical solution of ^{234}U due to the radiation damage of the crystal lattice caused by the decay of the parent ^{238}U and subsequent decays (Rosholt et al., 1963), and electron stripping during the decay process such that ^{234}U is more likely to be in the more soluble U^{6+} state, facilitating the solution of this isotope by a surface etching process (Jurado Vargas et al., 1995). Therefore, the $^{234}U/^{238}U$ activity ratios of surface or ground waters usually exceed the secular equilibrium ratio of unity. The key factors in affecting the $^{234}U/^{238}U$ activity ratios are the relative rates of leaching, mechanical erosion and daughter half-life (Osmond and Ivanovich, 1992). Published $^{234}U/^{238}U$ ratios of global rivers range from 1.03 to 2.59, averaging 1.17 (Chabaux et al., 2001).

As the world's second largest river in terms of sediment transport (Milliman and Meade, 1983; Wang et al., 1986), Yellow River was notable for its high dissolved U concentrations (Palmer and Edmond, 1993; Chabaux et al., 2001). Liu (1988) measured 55 water samples collected throughout the Yellow River drainage basin by the trialkylphosphine oxide extraction-solid fluorometry method. The results showed a range between 1.07 and 28.84 μg/l with a mean value of 7.39 μg/l. In a section of the river near Lanzhou (see Fig. 1), Chen et al. (1995) obtained a U concentration range 2.16–15.9 μg/l by the fission-track method. Cheng and Zhang (1999) used a laser-fluorometry method to determine 56 water samples in the river main channel and obtained a mean value of 7.51 μg/l. In their study, the water samples were

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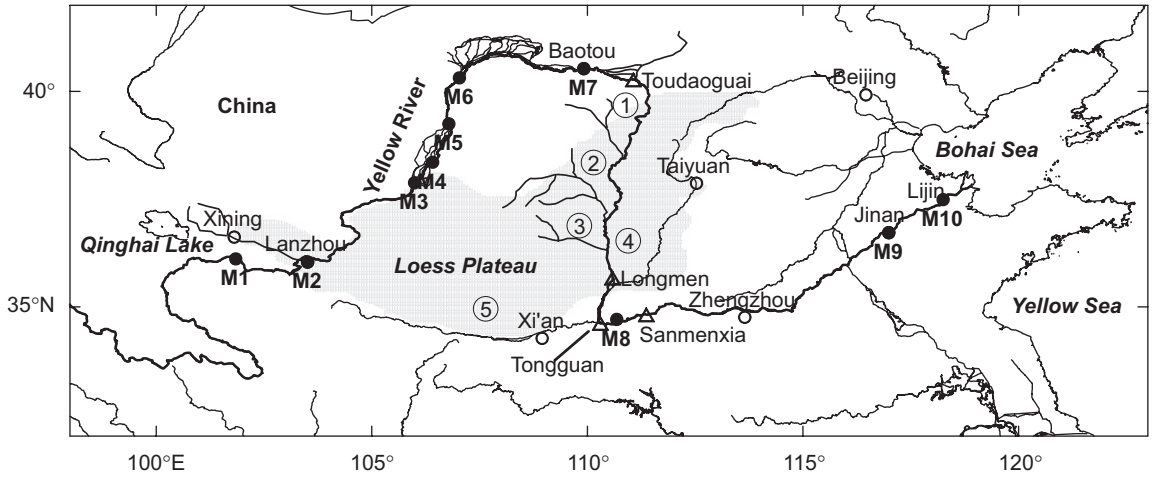

Fig. 1. Map of the Yellow River showing several of its main tributaries (① Kuye River; ② Wuding River; ③ Yan River; ④ Fen River; ⑤ Wei River) in the middle reaches and the sample sites (M1–M10) of this study. The shaded area represents the Chinese Loess Plateau (after Gu et al., 1997). ● Sample site; ○ city; △ hydrographic station.

unfiltered; they were obtained by decantation after settling of the suspended sediments contained therein. The methods of sample pretreatment were not described in the other two papers (Liu, 1988; Chen et al., 1995). Furthermore, the $^{234}U/^{238}U$ activity ratios were not measured in these three studies. Using thermal ionization mass spectrometry (TIMS), Chabaux et al. (2001) measured a filtered Yellow River water sample collected at Jinan (Fig. 1; mis-spelled as Jimau in Chabaux et al., 2001) and obtained a dissolved U concentration of 7.5 μg/l and a $^{234}U/^{238}U$ activity ratio of 1.318.

There has been no consensus on the origin of the high U concentration of the Yellow River. Palmer and Edmond (1993) attributed it to the dissolution of uraniferous organic-rich shales commonly intercalated with the evaporites in the upper reaches of the river. Zhou and Xu (1986) pointed to several possible sources, including U-rich underground brines leakage and weathering of uranium ore in the Yellow River drainage basin. Using alpha spectrometry, the present study measures uranium concentrations and, for the first time, the activity ratios in filtered waters and suspended particulate matters (SPM) from upper to lower reaches of the Yellow River main channel. A series of laboratory leaching and equilibration experiments was also conducted, together with the determination of the U content and activity ratio of the loess samples used in the experiments. Earlier, we reported the distribution of uranium in the Yellow River Plume/Estuary (Jiang et al., 2007). The main purpose of the present study lies in describing the uranium distribution and assessing the sources of dissolved uranium in the main channel of Yellow River. The data of this work should also contribute to the evaluation of uranium budget in the world ocean.

2. Study area

The 5464 km-long Yellow River is sourced from the northern part of the Bayankala Mountains at an altitude of 4830 m in the Qinghai-Tibet Plateau, China (Huang et al., 1992; Fu, 1998). Its drainage basin lies in the latitudinal and longitudinal belts of 32–42°N and 95–120°E (Fig. 1). The river is well-known for its high sediment discharge (averaging 1.6×10^{12} kg/yr; Xi, 1996), comparable or second only to the 1.67×10^{12} kg/yr estimated for the Brahmaputra River (Milliman and Meade, 1983; Huang et al., 2005). However, the total suspended solids of Yellow River water (27.6 g/l on average) are much higher than those of the Brahmaputra River (1.72 g/l), due to the difference in water discharge: 5.8×10^{10} m^3/yr for the Yellow River vs. 97.1×10^{10} m^3/yr for the Brahmaputra River (Milliman and Meade, 1983; Huang et al., 2005).

The upper reaches of the Yellow River drain the northeastern part of the Qinghai-Tibet Plateau at above 3000–4000 m. They supply ca. 60% of the river discharge but ca. 10% of the sediment load. The surface strata of this area are composed of sandstone, dolomitic limestone and minor volcanic rocks (Yang et al., 1986). The Loess Plateau covers part of the upper reaches and most of the middle reaches (Fig. 1) with an area of $\sim 3 \times 10^5$ km^2, some 40% of the total Yellow River drainage basin (Zhang et al., 1990). The Plateau is characterized by fragile structure, low resistance to erosion, columnar jointing, poor vegetation and arid-semiarid climate with evapotranspiration much higher than precipitation (Zhang et al., 1990). Rain storms are common in the summer. These features make the Chinese Loess Plateau the most physically eroded region of the world (Huang et al., 1992) with a physical erosion rate about 75 times larger than the chemical weathering rate (Chen et al., 1984). Materials eroded from the Plateau contribute 90% of the river sediment load (Zhang et al., 1990; Huang et al., 1992). The suspended sediment concentration increases significantly from less than $5–10 \times 10^3$ mg/l in the upper stream to $30–35 \times 10^3$ mg/l in the Loess Plateau region (Zhang et al., 1995). In its lower reaches, the river traverses the fluvial plain of northern China at an altitude of 50–100 m, where 33% of the suspended sediments settle to the river bed (Long and Xiong, 1981; Milliman and Meade, 1983). Due to the heavy sedimentation (ca. 0.5×10^{12} kg/yr), most of the river bed sits 5–10 m higher than the alluvial beyond the river banks (Huang et al., 1992).

3. Sampling and methods

3.1. Samples from the main channel

Samples M1–M10 from the Yellow River main channel (Fig. 1) were collected during four field trips between August 2005 and July 2006. The sampling locations were selected to be far away from tributary entries and any apparent sources of pollution. Surface water samples of 15–20 l were taken along the river bank using an acid-cleaned 3 l plastic pail and then poured into acid-cleaned plastic containers and transported back to the laboratory within 2–5 days.

The samples were filtered through 0.45 μm filters immediately upon their arrival at the laboratory. Uranium analyses followed