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博士学位论文

镭同位素示踪胶州湾海底地下水排泄

Tracing Submarine Groundwater Discharge into Jiaozhou Bay by radium isotopes

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摘要

本文依托国家自然科学基金项目“多种方法研究胶州湾海底地下水排泄（41072174）”，采用天然镭同位素（224Ra和226Ra）作为示踪剂，探讨沉积物中镭的扩散和解吸规律，分析胶州湾及其周边水体中224Ra和226Ra的时空分布特征；用224Ra和226Ra比值估算水体的表观年龄；通过三端元混合模型和镭质量平衡模型，评价胶州湾海底地下水排泄（SGD）及其输送的营养盐通量。

通过沉积物室内培养实验，发现同一沉积物在不同盐度水体条件下，随着水体盐度的逐渐增加，224Ra、226Ra解吸活度呈增加趋势，说明盐度越高，沉积物越容易解吸释放Ra到水中。不同粒径沉积物在同一水环境条件下，粒径为125μm￣250μm之间时，224Ra、226Ra解吸活度非常接近；粒径＞2000μm的224Ra、226Ra解吸活度略高，可能是这个粒级范围的沉积物空隙较大，间隙水向上覆水体扩散通道比较通畅，可以使较多的镭短时间内扩散到水体中；而粒级＜125μm的224Ra、226Ra解吸活度最大，其原因是沉积物颗粒越小，其比表面积越大，吸附的镭同位素越多，因而遇到咸水解吸出来的镭同位素也越多。通过实验获得胶州湾表层沉积物中224Ra的平均扩散通量为0.04 Bq•m-2h-1、226Ra的平均扩散通量为0.002 Bq•m-2h-1；沉积物再悬浮颗粒解吸的224Ra平均活度为 10.6Bq•kg-1、226Ra平均活度为9.8 Bq•kg-1。

2011年9-10月胶州湾周边区域地下水中224Ra活度平均为8.54 Bq•m-3，226Ra活度平均为2.23 Bq•m-3；2012年4-5月地下水中224Ra平均活度为8.04 Bq•m-3，226Ra平均活度为2.51 Bq•m-3。2011年9-10月胶州湾周边河水中224Ra活度平均值为2.34 Bq•m-3，226Ra活度平均值为1.08 Bq•m-3；2012年4-5月河水中224Ra平均值为2.44 Bq•m-3，226Ra平均值为1.27 Bq•m-3。2011年9-10月胶州湾湾内海水中224Ra活度平均值为3.83 Bq•m-3，226Ra活度平均值为2.67 Bq•m-3；2012年4-5月224Ra平均值为3.85 Bq•m-3，226Ra平均值为2.63 Bq•m-3。

基于224Ra/226Ra比值估算水体年龄，胶州湾表层水体年龄呈现较好的规律性，由湾顶到湾口，从北到南，呈逐渐增大趋势，距离湾顶越远水体年龄越大，符合水体

循环更新的实际情况。

通过三端元线性混合模型估算，2011年9-10月，胶州湾表层水体中地下水混合比率为11.6%，海水混合比率为78.6%，河水混合比率为9.9%； 2012年4-5月，胶州湾表层水体中地下水混合比率为6.5%，海水混合比率为86.8%，河水混合比率为6.7%。在海水、河水、地下水三个端元的混合比例基础上，估算出胶州湾2011年9-10月和2012年4-5月的SGD输入量分别为7.53×106m3•d-1和 4.24×106m3•d-1。

通过构建胶州湾水体中224Ra和226Ra质量平衡模型，计算得出2011年9-10月胶州湾SGD输入量为21.01×106 m3•d-1（根据224Ra质量平衡模型），15.17×106 m3•d-1（根据226Ra质量平衡模型）；2012年4-5月胶州湾SGD输入量为11.33×106 m3•d-1 （根据224Ra质量平衡模型），6.53×106 m3•d-1 （根据226Ra质量平衡模型）。根据水文地质数值模型计算的胶州湾陆源地下淡水排泄量（2011年9-10月，2.16×104 m3•d-1；2012年4-5月，1.72×104 m3•d-1），发现陆源地下淡水排泄量仅占总SGD的0.3%，而再循环海水排泄量占了99.7%左右。

以总SGD量计算其输入的营养盐量，2011年9-10月，SGD输入胶州湾的溶解性无机氮（DIN）为1190.77×104 mol•d-1、溶解性硅酸盐为302.98×104 mol•d-1、活性磷酸盐为0.20×104 mol•d-1；2012年4-5月，SGD输入胶州湾的溶解性无机氮为534.92×104 mol•d-1、溶解性硅酸盐为175.18×104 mol•d-1、活性磷酸盐为7.95×104mol•d-1。与河流输入的营养盐相比较，同一时期二者输入的溶解性无机氮很接近；输入的活性硅酸盐2011年9-10月比较接近，2012年4-5月则相差近5倍；输入的活性磷酸盐变化比较大，2011年9-10是河流输入的大，而2012年4-5月是SGD输入的大。总体来看，SGD输入的营养盐基本与河流输入的营养盐相当，可见SGD是胶州湾营养盐输入的一个重要途径。

关键词：海底地下水排泄（SGD）；224Ra；226Ra；水体年龄；营养盐；胶州湾

Abstract

This paper is supported by the project of using multi-methods research submarine groundwater discharge (SGD) in Jiaozhou Bay (41072174), which belongs to the National Natural Science Foundation of China. The purposes are using natural radium isotopes (224Ra and 226Ra) as a tracer to explore the diffusion and desorption of radium from sediment, and analyze spatial and temporal characteristics of 224Ra and 226Ra around Jiaozhou Bay; estimating the apparent age of Jiaozhou bay water based on 224Ra and 226Ra ratio; evaluating submarine groundwater discharge and its nutrient inputs through three end-members mixing model and radium mass-balance model.

By the sediment incubation experiments, we found that in different salinity water conditions, the desorption activities of 224Ra and 226Ra from the same sediment tended to increase with the gradual increase of salinity, which indicated that the higher salinity, radium isotopes from sediment more easily desorbed into the water.In the same water condition, when the particle sizes of sediments were between 125μm ~ 250μm, 224Ra and 226Ra desorption activities were very close. When the particle sizes of sediments were greater than 2000μm, 224Ra and 226Ra desorption activities were slightly higher. Because the interval of sediments in these particle sizes was so larger that the diffusion channels were relatively smooth between interstitial water and overlying water, there were more radium isotopes diffusing into the overlying water in short time. When the particle sizes of sediments were less than 125μm, 224Ra and 226Ra desorption activities were maximum. Due to the smaller particle sizes of sediment which led to the larger specific surface area, there were more radium isotopes adsorbed on the particles and then desorbed with mixing the salt water.Through the experiments, average diffusion flux of 224Ra from the surface sediments in Jiaozhou Bay was 0.04 Bq•m-2h-1, 226Ra was 0.002 Bq•m-2h-1; average desorption

activity of 224Ra from the resuspended particles of sediments was 10.6Bq•kg-1, 226Ra was9.8 Bq•kg-1.

The results showed that during September to October in 2011, average activities of 224Ra and 226Ra in groundwater around Jiaozhou Bay were 8.54 Bq•m-3 and 2.23 Bq•m-3; during April to May in 2012, average activities of 224Ra and 226Ra in groundwater around Jiaozhou Bay were 8.04 Bq•m-3 and 2.51 Bq•m-3. During September to October in 2011, average activities of 224Ra and 226Ra in river water around Jiaozhou Bay were 2.34 Bq•m-3 and 1.08 Bq•m-3; during April to May in 2012, average activities of 224Ra and 226Ra in river water around Jiaozhou Bay were 2.44 Bq•m-3 and 1.27 Bq•m-3. During September to October in 2011, average activities of 224Ra and 226Ra in bay water Jiaozhou Bay were 3.83 Bq•m-3 and 2.67 Bq•m-3; during April to May in 2012, average activities of 224Ra and 226Ra in bay water around Jiaozhou Bay were 3.85Bq•m-3 and 2.63 Bq•m-3.

Via 224Ra/226Ra model, the apparent ages of Jiaozhou Bay water were estimated and presented relatively consistent patterns that ages were gradually increasing from top of bay to the mouth of the bay and from north to south. With the increasing distance from the top of the bay the ages of water became older, which was in line with the actual situation of the water cycle and update.

Based on the three end-member mixing model, the results showed that during September to October in 2011, the mixing ratio of groundwater, seawater and river water was 11.6%,78.6% and 9.9%; during April to May in 2012, the mixing ratio of groundwater, seawater and river water was 6.5%,86.8% and 9.6.79%. Then the fluxes of SGD were estimated to be 7.53×106m3•d-1 and 4.24×106m3•d-1 during September to October in 2011 and April to May in 2012, respectively.

Through the radium (224Ra and 226Ra) mass balance model, the results showed

that during September to October in 2011, the fluxes of SGD were 21.01×106 m3•d-1(based on 224Ra mass balance) and 15.17×106 m3•d-1(based on 224Ra mass balance); during April to May in 2012, the fluxes of SGD were 11.33×106 m3•d-1(based on 224Ra mass balance) and 6.53×106 m3•d-1(based on 224Ra mass balance). The fluxes of submarine fresh groundwater discharge were 2.16×104 m3•d-1(September to October in 2011) and 1.72×104 m3•d-1(April to May in 2012) based on hydrogeological numerical model. The fluxes of submarine fresh groundwater discharge accounted for only 0.3% of the total SGD, while recirculated saline groundwater discharge accounted for 99.7%.

The fluxes of nutrients input were calculated by the total SGD fluxes. During September to October in 2011, the fluxes of dissolved inorganic nitrogen (DIN), soluble silicate and reactive phosphate were 1190.77×104 mol•d-1, 302.98×104 mol•d-1and 0.20×104 mol•d-1. During April to May in 2012, the fluxes of dissolved inorganic nitrogen (DIN), soluble silicate and reactive phosphate were 534.92×104 mol•d-1, 175.18×104 mol•d-1and 7.95×104 mol•d-1. Compared with the nutrients inputs of river, both dissolved inorganic nitrogen inputs were very closed in the same period; reactive silicate inputs were closed during September to October in 2011, while during April to May in 2012 the inputs had a difference of nearly five-fold; reactive phosphate inputs had large changes, the inputs of river was bigger during September to October in 2011, but the inputs of SGD were bigger during April to May in 2012. Overall, nutrients inputs of SGD and river were roughly equal, so the nutrients inputs of SGD is an important way for nutrients inputs in Jiaozhou By.

Keywords: Submarine groundwater dischargeSGD224Ra226Raapparent age of waternutrientsJiaozhou Bay

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