

⁹⁵Zr 在鱼-海水-底泥模拟生态系统中的迁移与分布

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摘要:采用同位素示踪技术研究了⁹⁵Zr 在鱼-海水-底泥模拟系统中的迁移与分布。旨在探明海洋鱼类对⁹⁵Zr 的吸收以及⁹⁵Zr 在海水生态系统中的行为特性。结果显示:(1)海水中的⁹⁵Zr 比活度随时间呈快速下降, 降至一半所需的时间仅为 0.2d; 底泥吸附的⁹⁵Zr 主要集中在表层(0~2.5cm), 至试验后期(13~20d)其量占总量的 99.5%以上, 表明来自于海水中的⁹⁵Zr 被表层底泥吸附, 不易随渗流水向下迁移, 底泥对⁹⁵Zr 有强烈的吸附作用。(2)中华乌塘鳢从水体中摄入和吸收⁹⁵Zr 的主要器官是肠胃道(内脏), 与水体直接接触而吸附、吸收的⁹⁵Zr 主要集中在鱼鳃和鱼鳍中。肉、骨、肝脏和鱼卵中的比活度均比较低(略高于本底水平), 表明通过肠胃道吸收和鳃、鳍等组织吸附、吸收的⁹⁵Zr 不易向肉、骨、肝脏和鱼卵等内部组织运输。(3)运用封闭 3 分室模型原理对实验数据进行拟合得: 鱼体中的比活度 $C_f = 0.05 - 6.36e^{-3.4274t} + 6.61e^{-0.2682t}$, 海水中的比活度 $C_w = 215.42e^{-3.4274t} + 2.00e^{-0.2682t}$, 底泥中的比活度 $C_s = 143.86 + 14.83e^{-3.4274t} - 158.69e^{-0.2682t}$; 方差分析结果表明各回归方程较好地反映了⁹⁵Zr 在鱼-海水-底泥生态系统中的消长动态。

关键词:⁹⁵Zr; 海水生态系统; 中华乌塘鳢; 迁移与分布; 放射生态学

Migration and distribution of ⁹⁵Zr in a simulated marine fishes-seawater-sediment ecosystem

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Abstract: The isotope tracer technique is used to study the dynamics of migration and distribution of ⁹⁵Zr in a simulated marine fishes-seawater-sediment system, and its dynamics is described mathematically using a compartment model with a non-linear fitting method. The objectives of this study are (i) to clarify the behavior trait of ⁹⁵Zr in marine ecosystem; (ii) to evaluate accumulation and distribution of ⁹⁵Zr in marine fishes; and (iii) to simulate the dynamics of ⁹⁵Zr in the marine fishes-seawater-sediment system based on a closed three compartments system model.

Three glass pools with dimensions of 40cm×50cm×40cm were constructed. 12.5 kg of sea sediment was filled into each pool. And sediment depth was about 5 cm. The pools were flooded with 42l seawater. Eleven mL of ⁹⁵ZrF₄ with a specific activity of 9.02×10^5 Bq/ml was introduced into the seawater after one

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week. The seawater was stirred gently to get a homogeneous distribution of radionuclide. The radionuclide specific activity in seawater was 216.9Bq/ml. Seven marine fishes (*Bostichthys sinensis*) were put into each pool. Seawater was added with intervals of 3 days in order to maintain a constant height of seawater.

The samples were collected at the elapsed time of 0, 25, 1, 2, 4, 8, 13 and 20 days. Four 5 ml aliquotes of seawater were randomly collected from each pool, and disposable plastic cups were filled with them (20 ml) for activity measurements. A marine fishes was taken from each pool. The fish was divided into fin, viscera, liver, gill, skin, meat, bone, head and roe, and each part was weighed. They were afterwards cut into smaller pieces. Twenty grams samples from each part were put into the disposable plastic cups for measurements.

In the meantime, two sediment columns were collected from each pool using a sediment sampler. Each sediment column was sectioned into two equal parts, and each part was smashed and mixed thoroughly. Afterwards 20 g of sediment samples were put into plastic cups for measurements. Every sample had 3 replications.

The ⁹⁵Zr emits β and γ particles when it decays. Those were measured with a multi-channel γ spectrometer (model BH 1224, Beijing Nuclear Instrumentation Factory). The counting error was controlled to be lower than 5%. The counting data were calibrated with counting efficiency, dead time, disintegration and any other factors.

The results showed: The specific activity of ⁹⁵Zr in seawater decreased rapidly with time due to precipitation, adsorption to sediment and uptake by marine fishes, and its half life was only 0.2 day. Most of ⁹⁵Zr in sediment was found concentrated in the surface layer, and the specific activity of ⁹⁵Zr in lower layer (2.5~5cm from surface) were almost in background level. About 99.5% of ⁹⁵Zr in sediment were found in surface layer in later stage (from 13th day to 20th day). It was indicated that ⁹⁵Zr in seawater could not readily move downwards with percolating water after remained in surface sediment. The main organ of *Bostichthys sinensis* to uptake and absorb ⁹⁵Zr was intestines and stomach (viscera). ⁹⁵Zr that absorbed by direct contact with seawater concentrated mainly on the gill and fin. The specific activity of ⁹⁵Zr in meat, bone, liver and roe was relatively lower, which were only slightly greater than the background level. It was indicated that ⁹⁵Zr remained in intestines, stomach, gill and fin could not readily transport to inner organ such as meat, bone, liver and roe.

A closed three-compartment system model was applied to imitate the experimental data. For dynamics of specific activity in whole fish, seawater and sediment, it could be described with the following exponential regression equations respectively: the specific activity of fish $C_f = 0.05 - 6.36e^{-3.4274t} + 6.61e^{-0.2682t}$, and seawater $C_w = 215.42e^{-3.4274t} + 2.00e^{-0.2682t}$, and sediment $C_s = 143.86 + 14.83e^{-3.4274t} - 158.69e^{-0.2682t}$ were gained. The results of analysis of variance showed that each regression equation can described the dynamics of accumulation and disappearance of ⁹⁵Zr in fishes-seawater-sediment system preferably.

Key words: ⁹⁵Zr; marine ecosystem; *Bostichthys sinensis*; migration and distribution; radioecology

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为了实现核电的持续发展和保持高质量环境的统一,研究核电站放射性排放物在生物体和生态系统中的行为特性是一个相当活跃的研究领域。有关生物体对核电站放射性排放物⁸⁵Sr、¹³⁷Cs、⁶⁰Co等核素的吸收及其在生态系统中的行为,尤其是迁移和累积等行为特性前人已作了较多的研究^[1~8]。但对⁹⁵Zr在生物体和生态系中,特别是行为动力学方面的研究鲜见报道,有关⁹⁵Zr的研究报道仅见切尔诺贝尔核电站附近地区及事故后世界各地对主要放射性核素(⁹⁵Zr是其中之一)的环境监测^[9~11]。本试验采用同位素示踪技

术研究⁹⁵Zr模拟污染物在中华乌塘鳢体内的分布及其在鱼-海水-底泥系统中的迁移与消长动态,运用分室模型和非线性拟合方法建立其动态行为的数学模型,并运用数理分析方法用实验数据估计拟合方程的误差和有效性。旨在探明⁹⁵Zr在模拟海水生态系统中的行为特性,在中华乌塘鳢体内的迁移和积累,为评价⁹⁵Zr对生物体及生态环境的影响提供科学依据。

1 材料与方法

1.1 同位素、海泥、海水和海洋鱼类

(1)⁹⁵ZrO₂由中国原子能研究院同位素所提供,黑色粉末状固体,比活度为 2.284×10^8 Bq/g,放化纯度大于95%,使用前用氢氟酸^[12]将其转化为 1.23×10^7 Bq/ml(2000年6月20日)的⁹⁵ZrF₄溶液。吸取⁹⁵ZrF₄溶液1.1ml稀释至15ml,得比活度为 9.02×10^5 Bq/ml的⁹⁵Zr水溶液。(2)海洋底泥取自浙江宁波象山港,使用前经风干、粉碎、去除石块等杂物后30目过筛。主要理化性质为:pH(水浸)8.5,有机质8.0g/kg,粘粒13.9%,粉粒61.5%,砂粒24.6%。(3)海水采自宁波象山港,盐度为28,按2份海水加1份淡水的比例配成盐度为18.6混合海水待用。(4)中华乌塘鳢(*Bostriichthys sinensis*),一种广盐性浅海经济鱼类,每尾重约130~150g,由宁波大学海洋与水产系提供。试验前在20℃室温条件下驯养1周,以适应供试环境。

1.2 实验方法

在3只40cm×50cm×40cm玻璃缸内,各加入海泥12.5kg,土层厚度约5cm,同时每缸埋入20只Φ23mm×50mm塑料取土器,先用适量盐度为18.6的混合海水湿润海泥后,再加入混合海水至刻度线(总水量为42L)。静置1周后,于2000年6月28日每缸引入比活度为 9.02×10^5 Bq/ml(2000年6月20日)的⁹⁵Zr水溶液11ml,均匀滴注在供试玻璃缸中,并用玻璃棒谨慎搅拌,使之均匀分布,⁹⁵Zr引入时海水中的平均比活度约为216.9Bq/ml。同时迅即每缸采取水样 2×20 ml(共6个水样)。然后每缸放养中华乌塘鳢7尾。整个试验期间室温、水温控制在20±2℃,并用充气泵给水体充氧。每天早晚2次给鱼喂食虾肉若干,以保持正常生长。试验期间每隔3d加水1次以保持水体深度基本恒定,并于每次采样前1h加水至刻度线。

于⁹⁵Zr引入后的0.25、1、2、4、8、13和20d采样。每缸随机选取4处各吸取海水5ml,合并(共20ml)置于Φ75mm×110mm的一次性塑料测样杯中;每池随机选取鱼1尾,用清水冲洗后用吸水纸吸去表面水,称重后解剖为鳍、内脏、肝、鳃、皮、肉、骨、头和鱼卵(如果有的话)9个部位,分别称重、剪碎后取20g(量少的部位取全部)装入一次性塑料测样杯中;每池随机取底泥取土器2只,将底泥分为上下两层,将各层底泥充分拌匀后称20g分别装入测样杯中待测量。每种样品3个重复。

1.3 测量仪器及测量方法

⁹⁵Zr发射β和γ射线,半衰期64d,放射性活度测量采用BH1224型微机-多道一体化能谱仪(北京核仪器厂生产)测其γ射线。该谱仪配置倒置的Φ70mm NaI闪烁探头,安装在铅屏蔽室中。测样器皿采用自备的Φ75mm×110mm的一次性塑料测样杯,将其置于倒置的闪烁探头上面,并用自制的定位装置固定测量位置,以保证所有样品测量几何位置的一致性。探头工作电压623V,阈值0.28。选取一个谱峰(240~300道)进行计数测量,测量结果经探测效率、死时间、衰变等校正后换算成样品的放射性比活度,测量误差控制在5%以内。所有样品在采样当天完成测量,以减少样品由于水份蒸发而造成的误差。

1.4 数学建模及方差分析

采用封闭三分室模型和非线性回归分析方法对实验数据进行⁹⁵Zr在鱼/海水/底泥系统中消长动态的曲线拟合,建立动态拟合方程;并对拟合方程进行方差分析,确定在95%置信水平下的置信区间。

2 结果与讨论

2.1 ⁹⁵Zr在水体和底泥中的变化动态

水体和底泥上下两层中⁹⁵Zr比活度随时间的变化动态见表1。表中数据显示水体中的⁹⁵Zr比活度随时间呈快速下降,至第2天已下降至初始比活度的0.75%,其原因是水体中的⁹⁵Zr一方面由于沉降、底泥吸附而滞留在表层底泥中,另方面被鱼体摄入以及由于吸附作用而粘附于鱼体表面(表2),因而致使水体中的⁹⁵Zr比活度快速下降;底泥中的⁹⁵Zr主要集中在上层(0~2.5cm),且随时间呈逐渐增加,下层(2.5~5cm)较少,其比活度总体上接近于本底水平,随时间呈缓慢增加趋势,至试验后期(13~20d)表层土壤中⁹⁵Zr的

量占总量的99.57%~99.68%。表明来自于水体中的⁹⁵Zr被上层底泥吸附,不易随渗流水向下迁移。

2.2 ⁹⁵Zr在鱼体中的积累与分布动态

⁹⁵Zr在鱼体各部位中的分布动态见表2。表中结果显示:中华乌塘鳢从水体中摄入和吸收⁹⁵Zr的主要器官是肠胃道(内脏),与水体直接接触而吸附、吸收的⁹⁵Zr主要集中在鱼鳃和鱼鳍中,此3部位⁹⁵Zr的比活度至0.25d已达最大值,而后随时间呈逐渐下降趋势,结合水体的比活度变化动态,可以认为起初该3部位从水中快速摄入、吸附⁹⁵Zr而导致其比活度快速上升,而后随水体中比活度的降低,其排泄、解吸速率超过了摄入、吸附速率,因而其比活度又呈逐渐下降;从鱼鳃中的比活度数值看,其在0.25、1、3d时的比活度分别是相应时间水中比活度的3.62、24.40倍和8.62倍,表明鱼鳃对水体中的

⁹⁵Zr具有强烈的吸附和固定作用,这显然与鱼鳃的结构、功能和其巨大的表面积有关;肉、骨、肝脏和鱼卵的比活度均比较低(略高于本底水平),表明通过肠胃道吸收和鳃、鳍等组织吸附、吸收的⁹⁵Zr不易向肉、骨、肝脏和鱼卵等内部组织转运,其原因主要与锆的生物学特性(锆不是生命必须元素,在生物系统中的流动性低)有关;鱼皮对⁹⁵Zr的吸附、吸收能力较鱼鳍弱,且随水体中⁹⁵Zr比活度的降低也呈逐渐下降趋势;鱼头的组织结构比较复杂,既有骨、肉等内部组织,又有暴露于水体的外表皮和口腔组织,由于骨、肉等内部组织中的⁹⁵Zr比活度非常低,因此鱼头中的⁹⁵Zr主要来源于与水体直接接触的外部组织的吸附和吸收,其比活度在1d时达峰值(2.21Bq/g)后也随时间逐渐下降。

表1 ⁹⁵Zr在水体和底泥中的比活度变化动态

Table 1 Dynamic change of ⁹⁵Zr specific activity in the seawater and sediment

时间 Time(d)	各土层中的比活度 Specific activity in soil profile(Bq/g)		海水中的比活度 Specific activity in seawater (Bq/ml)
	0~2.5cm	2.5~5cm	
0	0.00	0.00	217.42
0.25	4.15	0.01	44.28
1	18.54	0.12	3.68
2	20.48	0.29	1.62
4	91.74	0.15	0.50
8	169.90	0.80	0.22
13	278.83	1.19	0.06
20	285.75	0.93	0.01

表2 ⁹⁵Zr在鱼体中的比活度分布动态(Bq/g)

Table 2 Dynamic distribution of ⁹⁵Zr specific activity in the fish

时间 Time(d)	鱼体(鲜重) Fish(fresh weight)							
	鳍 Fin	内脏 Viscera	肝 Liver	鳃 Gill	皮 Skin	肉 Meat	骨 Bone	头 Head
0.25	8.68	7.24	0.66	160.42	1.27	0.01	0.05	1.86
1	4.91	4.62	0.17	89.79	0.96	0.01	0.05	2.21
2	4.05	1.86	0.08	13.97	0.53	0.03	0.06	0.52
4	2.87	2.22	0.16	10.18	0.38	0.00	0.11	0.56
8	1.13	1.22	0.13	6.73	0.24	0.00	0.09	0.37
13	0.27	0.69	0.12	6.38	0.24	0.02	0.17	0.26
20	0.30	0.45	0.04	3.54	0.21	0.03	0.15	0.16

2.3 ⁹⁵Zr在鱼-海水-底泥系统中的迁移、消长规律

⁹⁵Zr在鱼-海水-底泥系统中的消长行为可运用具有相互交换的封闭3分室模型描述,如图1所示。令各分室中⁹⁵Zr的量对时间的变化率服从1级速率过程^[13]。若以 q_f 、 C_f 、 m_f 、 q_w 、 C_w 、 m_w 和 q_s 、 C_s 、 m_s 分别表示鱼、海水和底泥中⁹⁵Zr的总活度、比活度和质量; k_{ws} 、 k_{fw} 、 k_{sw} 、 k_{sw} 、 k_{sf} 和 k_{fs} 分别为各分室间的转移速率常数。则各分室中⁹⁵Zr总活度对时间的变化率为:

$$\frac{dq_w}{dt} = k_{ws}q_s + k_{fw}q_f - (k_{ws} + k_{fw})q_w$$

$$\frac{dq_s}{dt} = k_{ws}q_w + k_{fs}q_f - (k_{sf} + k_{ws})q_s$$

$$\frac{dq_f}{dt} = k_{sf}q_s + k_{fw}q_w - (k_{sf} + k_{fw})q_f$$

解此微分方程组,注意到 $q_s(0)=0$, $q_f(0)=0$, $q_f=m_fC_f$, $q_w=m_wC_w$, $q_s=m_sC_s$,并令:

$$k_1 = k_{ws} + k_{fw} \quad k_2 = k_{sw} + k_{sf} \quad k_3 = k_{fs} + k_{fw} \quad b = k_1 + k_2 + k_3$$

$$\begin{aligned}
 c &= k_1k_2 + k_2k_3 + k_1k_3 - k_{fw}k_{wf} - k_{sw}k_{ws} - k_{fs}k_{sf} \quad d = k_2 + k_3 \quad f = k_2k_3 - k_{fs}k_{sf} \\
 g &= k_{fs} + k_{fw} + \frac{k_{fw}k_{wf}}{k_{ws}} \quad h = k_{sw} + k_{sf} + \frac{k_{ws}k_{sf}}{k_{fw}}
 \end{aligned}$$

$\gamma = \frac{b - \sqrt{b^2 - 4c}}{2} \quad \delta = \frac{b + \sqrt{b^2 - 4c}}{2}$

便得：

$$\begin{aligned}
 C_f(t) &= \frac{m_w C_w(0) k_{wf}}{m_f} \left[\frac{h}{\gamma \delta} + \frac{h - \gamma}{\gamma(\gamma - \delta)} e^{-\gamma t} + \frac{h - \delta}{\delta(\delta - \gamma)} e^{-\delta t} \right] \\
 C_w(t) &= C_w(0) \left[\frac{f}{\gamma \delta} + \frac{\gamma^2 - d\gamma + f}{\gamma(\gamma - \delta)} e^{-\gamma t} + \frac{\delta^2 - d\delta + f}{\delta(\delta - \gamma)} e^{-\delta t} \right] \\
 C_s(t) &= \frac{m_s C_s(0) k_{ws}}{m_s} \left[\frac{g}{\gamma \delta} + \frac{g - \gamma}{\gamma(\gamma - \delta)} e^{-\gamma t} + \frac{g - \delta}{\delta(\delta - \gamma)} e^{-\delta t} \right]
 \end{aligned}$$

根据上述推导结果,表明鱼、海水和底泥分室中的⁹⁵Zr 比活度服从多项指数规律,对表 3 数据运用计算机进行指数回归分析^[14],得到各分室⁹⁵Zr 比活度的消长动态拟合方程式(列于表 4);对拟合方程进行方差分析^[15],计算在 95% 置信度下的置信区间,结果列于表 4。从各拟合方程式与实验值的标准误差 σ 看(其值分别为 1.69Bq/g、20.12Bq/ml 和 29.70Bq/g),与鱼体峰值比活度、海水起始比活度和底泥平衡比活度的比率分别为 28.63%、9.25% 和 20.74%,在合理的偏差范围内,表明拟合方程较好地反应了⁹⁵Zr 在鱼-海水-底泥系统中的消长动态。各分室中⁹⁵Zr 比活度的变化动态(实验值和拟合曲线)如图 2 所示,可见,海水中⁹⁵Zr 的比活度随时间的延长,逐渐趋向于 0;鱼体中的比活度随时间的延长下降速率逐渐趋缓,可以推测在经过较长一段时间以后,会达到动态平衡而趋于稳定;结合上述两分室的动态变化及底泥中⁹⁵Zr 的变化动态,土壤中⁹⁵Zr 的比活度也会逐渐达到动态平衡而趋于稳定。

3 结论

(1)从海水中引入的⁹⁵Zr 由于沉降、底泥吸附、鱼体吸收和吸附其比活度随时间呈快速下降,至第 2d 已下降至初始比活度的 0.75%;底泥中的⁹⁵Zr 主要集中在表层(0~2.5cm),且随时间呈逐渐增加,至试验后期(13d 至 20d)其量占底泥中总量的 99.5%以上。表明来自于海水中的⁹⁵Zr 被表层底泥吸附,不易随渗流水向下迁移,海泥对⁹⁵Zr 有强烈的吸附作用。

(2)中华乌塘鳢从水体中摄入和吸收⁹⁵Zr 的主要器官是肠胃道(内脏),与水体直接接触而吸附、吸收的⁹⁵Zr 主要集中在鱼鳃和鱼鳍中,其中鱼鳃中的⁹⁵Zr 比活度最高,超过了相应时间水中比活度,表明鱼鳃对水体中的⁹⁵Zr 具有强烈的吸附和固定作用。

(3)肉、骨、肝脏和鱼卵中的比活度均比较低(略高于本底水平),表明通过肠胃道吸收和鳃、鳍等组织吸附、吸收的⁹⁵Zr 不易向肉、骨、肝脏和鱼卵等内部组织运输。

(4)采用分室模型和非线性回归方法在计算机上进行⁹⁵Zr 在鱼-海水-底泥系统中的消长动态拟合,经方差分析,表明各回归方程较好地反应了⁹⁵Zr 在鱼-海水-底泥系统中的消长动态。

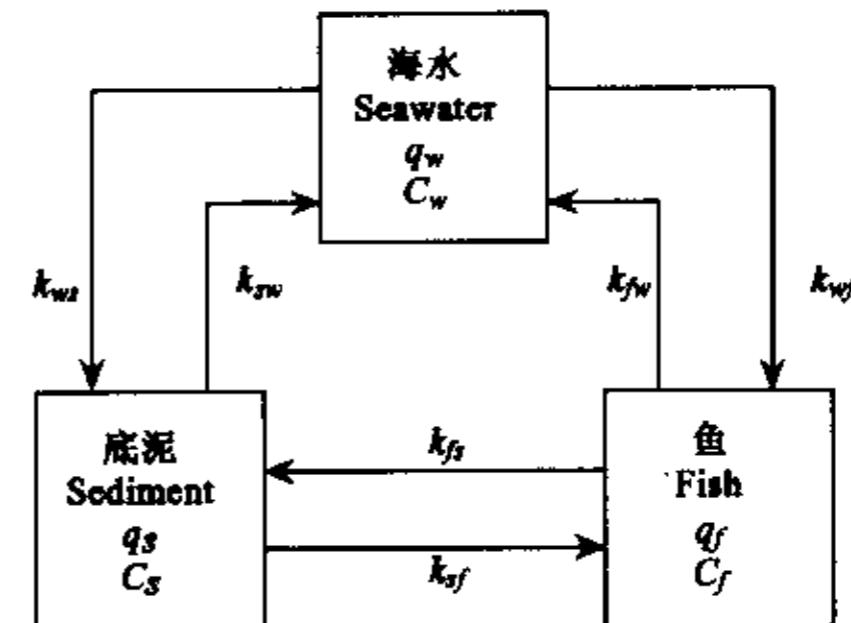


图 1 具有相互交换的封闭 3 分室模型

Fig. 1 The closed three-compartment system with interchange

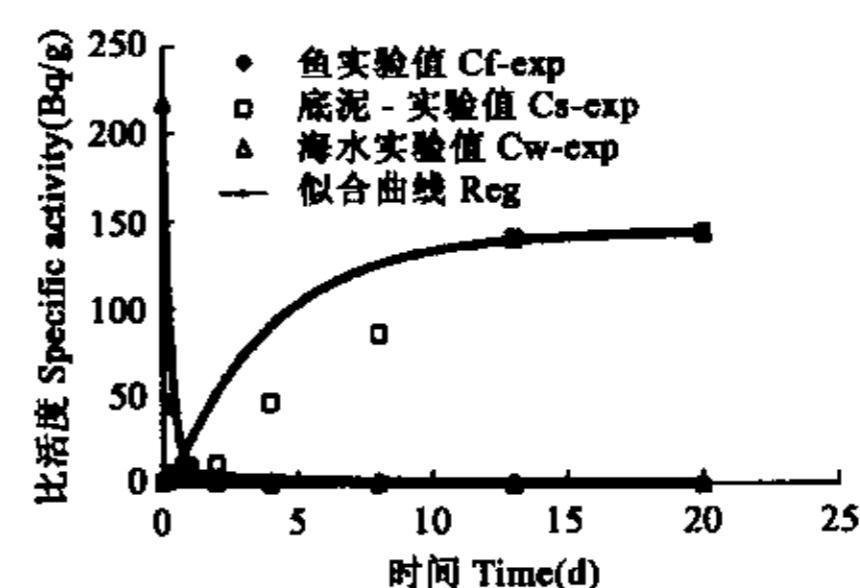


图 2 各分室的实验值与相应的拟合曲线

Fig. 2. The experimental data and suitability of fitting curves of each compartment
Where, reg. means exponential regression curves; exp. means experimental data

表3 各分室中⁹⁵Zr比活度的变化动态(Bq/g)Table 3 Dynamic change of ⁹⁵Zr specific activity in each compartment

分室 Compartment	时间 Time(d)							
	0	0.25	1	2	4	8	13	20
鱼 Fish	0.00	5.92	2.99	1.37	0.94	0.60	0.36	0.31
海水 Seawater	217.42	44.28	3.68	1.62	0.50	0.22	0.06	0.01
海泥 Sediment	0.00	2.08	9.33	10.39	45.95	85.35	140.01	143.34

表4 鱼-海水-底泥系统中⁹⁵Zr的消长动态拟合方程Table 4 Fitting equation of accumulation and disappearance of ⁹⁵Zr in the marine fishes-seawater-sediment system

分室 Compartment	拟合方程 Fitting equations	置信水平(%) Confidence level	置信区间 Confidence interval
鱼 Fish	$C_f(t) = 0.05 - 6.36e^{-3.4274t} + 6.61e^{-0.2682t}$	95	$C_f \pm 3.38$
海水 Seawater	$C_w(t) = 215.42e^{-3.4274t} + 2.00e^{-0.2682t}$	95	$C_w \pm 40.24$
海泥 Sediment	$C_s(t) = 143.86 + 14.83e^{-0.3.4274t} - 158.69e^{-0.2682t}$	95	$C_s \pm 59.40$

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