

放射性核素⁹⁵Zr 在水生态系统中的运输

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摘要 采用模拟污染物的同位素示踪技术研究了⁹⁵Zr 在水生态系统中的迁移、消长和分配动态, 并应用库室模型确定了各体系的拟合方程。结果表明:⁹⁵Zr 进入水中后, 在水生态系统中发生沉淀或与其他离子进行络合或被水生生物吸收或被吸附等形式在系统中迁移和转化, 从而在系统各部分中分配和积累。在引入后的很短时间, 池水中⁹⁵Zr 的比活度迅速降至一定值后缓慢下降。底泥通过与⁹⁵Zr 进行离子交换, 富集了大量的⁹⁵Zr。水葫芦(凤眼莲)也可在短期内吸附大量的⁹⁵Zr。螺蛳(环棱螺)和鲫鱼(鲫)对⁹⁵Zr 的吸附能力较弱, 螺蛳肉中⁹⁵Zr 的富集率大于在壳中的富集率,⁹⁵Zr 在鱼体内的分布主要集中在内脏中。⁹⁵Zr 在系统各部分的量均受时间的影响。

关键词 ⁹⁵Zr 水生态系统 迁移与分布 分室模型 放射生态学

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Migration and distribution of radioactive nuclide Zirconium-95 in the aquatic ecosystem

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Abstract : The dynamics of transportation, accumulation, diminishing and distribution of ⁹⁵Zr in a simulated aquatic ecosystem was studied by isotope-tracer technique, and the fitting equation was established by application of a closed five-compartment model. The results showed that when ⁹⁵Zr was introduced into aquatic system, it was transported and transformed via depositing, complex with other ions, adsorption and absorption by aquatic living organisms, resulted in redistribution and accumulation in part of the organisms. After introduction, the specific activity of ⁹⁵Zr in the water decreased sharply to a certain value in a short time and then decreased slowly. The sediment accumulated a large amount of ⁹⁵Zr by ion exchange. The water hyacinth (*Eichhornia crassipes*) could also adsorb a large amount of ⁹⁵Zr in a short period of time. Snail (*Bellamya purificata*) and fish (*Carassius Auratus*) had a poor capacity of adsorbing ⁹⁵Zr. The amount of ⁹⁵Zr in the snail flesh was greater than that in the shell, and the distribution of ⁹⁵Zr in the fish was found mainly in the viscera. The amount of ⁹⁵Zr in individual compartment of the system was affected with time.

Three glass tanks with dimensions of 60cm 40cm 50cm were constructed. 8kg of paddy soil on powdery loam was filled into each pool. The tanks were flooded with 60l water. 10 ml of ⁹⁵ZrF₄ with a specific activity of 4.32 × 10⁵ Bq/ml was

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introduced into the water after one week. The water was stirred gently to get a homogeneous distribution of radionuclide. The radionuclide specific activity in water was 72.00Bq/ml. Fifteen water hyacinth (*Eichhorma corssipes* (Mart) Solms), and 30 snails (*Bellamya purificata*) and 15 fish (*Carassius anratus*) were put into each tank. Water was added with intervals of 3 days in order to maintain a constant height of water.

The samples were collected at time interval of 1, 2, 4, 6, 9, 15, 22, 29 and 34 days. Four 5ml aliquots of water were random collected from each pool, and disposable plastic cups were filled with them (10ml) for activity measurements. A fish was taken from each tank. The fish was divided into fin, viscera, liver, gill, skin, flesh, bone, head and roe, and each part was weighed. They were then cut into smaller pieces. Twenty gram samples from each part were put into the disposable plastic cups for measurements.

In the meantime, two sediment columns were collected from each tank using a sediment sampler. Each sediment column was sectioned into two equal parts, and each part was smashed and mixed thoroughly. Afterwards 20g 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 counts was calibrated with counting efficiency, diminish time, disintegration and other factors.

The results showed that the specific activity of ⁹⁵Zr in water decreased rapidly with time due to precipitation, adsorption to sediment and uptake by water calabashes, and snails and fishes. Most of ⁹⁵Zr in sediment was found concentrated in the surface layer. It was indicated that ⁹⁵Zr in water could not readily move downwards with percolating water before remained in surface sediment. The specific activity of ⁹⁵Zr in different parts of organisms in orders of root > leaf in water hyacinth and flesh > shell in the snails. The organ uptaking and adsorbing ⁹⁵Zr was mainly of intestines and stomach (iscera). ⁹⁵Zr absorbed by gill and fin was though a direct contact with water. The specific activity of ⁹⁵Zr in flesh, bone, liver and eggs was relatively lower, which were only a little higher 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 flesh, bone, liver and eggs. The specific activity of ⁹⁵Zr in different parts of fish was arranged in order of viscera > gill > fin > skin, fish scale > bone, head, eggs > flesh.

A closed five-compartment system model was applied to imitate the experimental data. For dynamics of specific activity in whole water, sediment, water hyacinth, snails, and fishes, it could be described with the following exponential regression equations respectively, the specific activity of water $C_1 = 654.3447e^{-2.2147t} + 0.3285e^{-0.006690t} + 0.1119e^{-0.0129t} + 0.1099e^{-0.1848t} + 0.0006e^{-0.3378t}$, and sediment $C_2 = 85.7971e^{-2.2147t} + 1.6024e^{-0.006690t} + 91.8122e^{-0.0129t} + 2.9428e^{-0.1848t} + 0.3845e^{-0.3378t}$, and water hyacinth $C_3 = (118995.7705e^{-2.2147t} + 119516.3543e^{-0.6690t} + 682.5725e^{-0.0129t} + 656.1561e^{-0.1848t} + 5.3418e^{-0.3378t})/m_3_0$, and snail $C_4 = (0198.6263e^{-2.2147t} + 304.2514e^{-0.6690t} + 679.3532e^{-0.0129t} + 155.6352e^{-0.1848t} + 9201.0370e^{-0.3378t})/m_4_0$, and fish $C_5 = (13794.5841e^{-2.2147t} + 4717.3065e^{-0.6690t} + 6210.0565e^{-0.0129t} + 212256.7591e^{-0.1848t} + 23.4206e^{-0.3378t})/m_5_0$ were gained. The ANOVA showed that each regression equation can described the dynamics of accumulation and diminishment of ⁹⁵Zr in the aquatic ecosystem preferably.

Key Words : ⁹⁵Zr ; aquatic ecosystem ; migration and distribution ; compartment model ; radioecology

为实现核电的可持续发展和保持高质量环境的完美统一,研究核电站放射性排放物在生态环境中的行为特性是一个相当活跃的学科领域。⁹⁵Zr 是核电站反应堆的主要裂变产物之一,是压水堆核电站的主要放射性液态排放物。有关⁹⁵Zr 的研究报道主要集中在核事故后的环境监测和评价上,而关于⁹⁵Zr 在生态系统中的吸附、分配和积累的研究也有一些报道^[~6]。本试验采用同位素示踪技术,从动力学角度研究了⁹⁵Zr 在淡水生态系中的吸附、迁移、积累和分布,并运用分室模型原理与非线性拟合方法建立其数学模型,以寻求⁹⁵Zr 在生

物体和生态环境中的行为规律,对⁹⁵Zr 在上述系统中行为规律进行定量表征,为其进入生物体和生态系统后的安全性评价提供科学依据。

1 材料与方法

1.1 实验材料

1.1.1 供试核素

⁹⁵ZrO₂ 由中国原子能研究院同位素所提供,为粉末状固体,放射性比活度为 1.142×10^8 Bq/g (001-03-19,下同),放化纯度大于 95%,使用前用氢氟酸^[1] 将其转化为 6.165×10^6 Bq/ml 的⁹⁵ZrF₄ 溶液。吸取⁹⁵ZrF₄ 溶液 1.0 ml 稀释至 100 ml,得浓度为 6.165×10^4 Bq/ml 的⁹⁵Zr 水溶液供试验用。

1.1.2 供试土壤

底泥为小粉土,取自浙江大学华家池校区实验农场。使用前经粉碎,去除石块、植物根系等杂物,再风干、粉碎后 30 目过筛。其主要理化参数参见文献^[1]。

1.1.3 供试水生生态系

由水、底泥及水生生物 (水葫芦 (*Eichhorma corssipes* (Mart) Solms)、螺蛳 (*Bellamya purificata*)、鲫鱼 (*Carassius anratus*)) 组成。

1.2 实验方法

在 3 只 60 cm × 40 cm × 50 cm 玻璃鱼缸内,各加入小粉土 8000 g 作底泥,土层厚度约 5 cm,每缸加水 60L,每缸埋入 φ1 cm × 5 cm 塑料取土器 12 只,静置 1 周后,每缸分别引入前述的⁹⁵ZrF₄ 水溶液 10.00ml,其比活度为 4.32×10^5 Bq/ml,故水中⁹⁵Zr 的初始比活度为 72.00 Bq/g。用玻璃棒谨慎搅拌使之均匀,即刻取水样 20.0 ml。然后放入水葫芦 15 颗/池,螺蛳 30 枚/池,鲫鱼 45 尾 (5 尾/池),每尾重约 130 ~ 220 g。试验期间,用充氧泵昼夜充氧,使水体溶解氧恒定,经常补充蒸发损失的水分,以保持试验系统中的水量。

分别于⁹⁵Zr 引入后的 1、2、4、6、9、15、22、29、34d 采样。每缸随机选取 4 处各取样 5ml,共采水样 20ml,每池随机选取水葫芦 1 颗,螺蛳 3 枚,鱼 1 尾 所采集的水葫芦分为根、葫芦、叶,螺蛳分为肉和壳,鱼用自来水冲洗后用吸水纸吸干表面水,称重后解剖,分解为鱼鳍、鳞、内脏、皮、鱼鳃、肉、骨、头 8 个部位 (或加鱼籽共 9 个部位),分别称重后,取适量置于测样杯中待测量。所采集的底泥样品称重后 (减去取土器的重量),分为上下两层,将各层底泥充分拌匀后,每个样品称取 20.00g,置于测样杯中待测量。

本实验于 2001 年 5 月 21 日至 6 月 24 日,在浙江大学华家池校区原子核学科学研究所示踪实验室中进行。

1.3 实验仪器及测量方法

所有样品均在 BH1224 型微机-多道一体化能谱仪上按文献^[1] 中的测量方法,测其放射性活度,测量结果经探测效率、衰变等校正后换算成样品的比活度,测量的相对标准偏差控制在 5% 以内。

2 结果与讨论

2.1 ⁹⁵Zr 在池水和底泥中的消长

池水及底泥中⁹⁵Zr 的比活度随时间变化情况见表 1。⁹⁵Zr 进入水中后,由于沉淀、络合以及被底泥、水生动植物的吸附、吸收,使得池水中⁹⁵Zr 的比活度迅速下降,底泥中⁹⁵Zr 的比活度迅速上升。1 d 后,池水中⁹⁵Zr 的比活度就只有 19.52B q/ml,仅为初始比活度 (2.00 Bq/g) 的 27.11%。底泥在 4 d 时达上层到最大值,4 d 后,部分与底泥结合并不紧密的⁹⁵Zr 开始解吸,使得底泥中⁹⁵Zr 的比活度又有所下降。随着时间的推移,水中锆离子与生态系中其他各组分中的离子交换已基本上达到一种动态平衡,底泥中⁹⁵Zr 开始向深层自然迁移,并通过络合、铁锰氧化物吸附等途径与底泥形成相对稳定的结合,致使底泥中可游离的⁹⁵Zr 离子也在急剧减少,呈现出维持在一个比活度水平上但略有减少的趋势^[8~11]。

2.2 ⁹⁵Zr 在水葫芦和螺蛳中的消长动态

水葫芦和螺蛳中⁹⁵Zr 的比活度与时间关系见表 2。在 1d 时,水葫芦中的⁹⁵Zr 已达到最大为 247.25Bq/g,

而后随着时间的推移,水葫芦中的⁹⁵Zr 可能由于解吸的缘故,其比活度有所下降。由于锆不是生物体必须的元素,因此只有少量的⁹⁵Zr 被水葫芦吸收并被稳固结合在其体内。随着螺蛳体内外水分的交换,大量的⁹⁵Zr 进入螺蛳体内,螺蛳中的比活度急剧上升,在达最大值后开始下降,随后,由于水中⁹⁵Zr 比活度急剧降低,螺蛳体内⁹⁵Zr 发生解吸,其比活度降至 15d 后基本不变。螺蛳肉中⁹⁵Zr 比活度显著高于螺蛳壳中的比活度。

表 1 水、底泥中⁹⁵Zr 比活度随时间的变化
Table 1 Change in ⁹⁵Zr specific activity in water and sediment with time

时间 Time (d)	水 Water (Bq/g)	底泥 (Bq/g) Sediment		
		上 Upper (0 ~ 2.5 cm)	下 Lower (2.5 ~ 5 cm)	平均 Average
1	19.52	188.70	1.25	95.61
2	12.23	288.32	3.04	147.21
4	8.22	304.21	4.43	156.55
6	5.60	229.64	7.34	122.16
9	4.46	243.05	9.60	131.13
15	2.35	185.85	12.52	105.45
22	0.59	196.64	13.37	111.69
29	0.31	218.47	14.23	123.46
34	0.23	209.05	16.20	119.67

表 2 水生生物水葫芦、螺蛳中⁹⁵Zr 比活度与时间关系
Table 2 Relationship between specific activity and time of ⁹⁵Zr in aquatic organisms in water hyacinth and snail

时间 Time (d)	水葫芦比活度 Water hyacinth of specific activity (Bq/g)				螺蛳比活度 Snail of specific activity		
	叶 Leaf	葫芦 Hyacinth	根 Root	整株平均 Whole plant average	肉 Flesh	壳 Shell	整枚螺蛳平均 Whole snail average
1	286.60	134.13	319.01	247.25	37.54	17.76	29.44
2	259.08	119.12	285.39	221.19	28.65	12.98	17.55
4	234.23	112.29	269.05	208.53	17.72	6.30	9.75
6	240.64	110.63	265.08	205.45	9.87	4.97	6.97
9	163.01	74.96	179.60	139.22	6.00	2.96	3.96
15	114.28	52.83	126.53	98.12	6.02	2.92	3.93
22	114.42	52.61	126.04	97.70	5.98	2.89	3.85
29	107.20	49.29	118.09	91.53	5.77	2.73	3.69
34	106.81	49.11	117.68	91.21	5.74	2.81	3.64

2.3 ⁹⁵Zr 在鱼体中的消长

⁹⁵Zr 在鱼体各部位中的消长分布动态见表 3,其中全鱼中⁹⁵Zr 比活度数据系由鱼体各部位比活度的质量

表 3 鱼中⁹⁵Zr 比活度与时间关系
Table 3 Relationship between specific activity and time of ⁹⁵Zr in aquatic organisms in fish

鱼体部位 Different parts of fish	采样时间 Sampling time (d)								
	1	2	4	6	9	15	22	29	34
鱼鳃 Fish gill (Bq/g)	8.36	7.97	6.20	4.93	3.05	1.41	1.93	0.92	0.89
鱼鳍 Fish fin (Bq/g)	1.35	9.60	1.41	0.43	0.62	0.68	0.29	0.19	0.17
鱼内脏 Fish viscera (Bq/g)	104.11	140.58	40.17	7.99	9.27	7.34	1.83	1.71	1.68
鱼皮 Fish skin (Bq/g)	0.56	0.99	0.19	0.07	0.04	0.05	0.09	0.03	0.01
鱼鳞 Fish scale (Bq/g)	0.22	0.71	0.17	0.28	1.17	0.16	0.11	0.09	0.07
鱼肉 Fish flesh (Bq/g)	0.03	0.06	0.01	0.01	0.01	0.00	0.00	0.00	0.00
鱼骨 Fish skeleton (Bq/g)	0.17	0.21	0.03	0.00	0.05	0.01	0.01	0.00	0.00
鱼头 Fish head (Bq/g)	0.23	0.26	0.15	0.12	0.16	0.09	0.08	0.02	0.03
鱼籽 Fish eggs (Bq/g)	0.35	0.00	0.07	0.00	0.03	0.01	0.00	0.04	0.00
全鱼 Whole fish (Bq/g)	6.85	11.52	3.38	0.35	1.41	0.77	0.20	0.14	0.07

加权计算获得。结果显示 鲫鱼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的⁹⁵Zr 主要集中在鱼鳃和鱼鳍中, 此 3 部位⁹⁵Zr 的比活度至 2d 已达最大值, 短期内随着水体中⁹⁵Zr 的减少而迅速减少, 而后随时间呈逐渐下降趋势, 4d 后减少的趋势趋于平缓, 大体趋势与螺蛳的消长情况相似。肉、骨和鱼籽的比活度均比较低 (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、骨和鱼籽等内部组织输运 鱼皮和鱼鳞对⁹⁵Zr 的吸附能力较鱼鳍弱, 且在第 2 天达最大值为 0.99Bq/g 和 0.71 Bq/g, 随后随水体中⁹⁵Zr 比活度的降低而呈逐渐下降趋势 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, 其中的⁹⁵Zr 主要是来源于与水体直接接触的外部组织的吸附和吸收, 其比活度与其他部位相比也是比较低的, 其变化动态与鱼皮和鱼鳞相似。

2.4 ⁹⁵Zr 在模拟水生生态系统中的迁移模型

本实验水生生态系统由池水、底泥、水葫芦、螺蛳、鱼构成, 由于该系统与外界不存在水分、离子交换, 因此可看作是一封闭五分室系统。根据实验的具体情况, 忽略一些次要过程, 可建立如图 1 所示的模型 [2-15]。

式中 k_{12} 、 k_{13} 、 k_{14} 、 k_{15} 为⁹⁵Zr 由水体向底泥、鱼、螺蛳、水葫芦的迁移速率, k_{21} 、 k_{24} 为⁹⁵Zr 由底泥向水体、螺蛳的迁移速率, k_{31} 为⁹⁵Zr 由鱼向水体的迁移速率, k_{42} 为⁹⁵Zr 由螺蛳向底泥的迁移速率, k_{51} 为⁹⁵Zr 由水葫芦向水体的迁移速率。据此, 各分室中⁹⁵Zr 量 q_1 、 q_2 、 q_3 、 q_4 、 q_5 对时间变化率为：

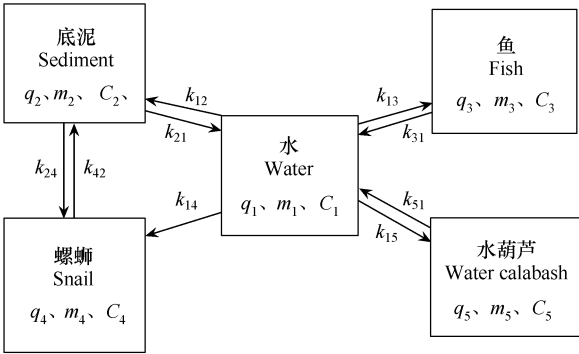


图 1 水生生态封闭五分室模型

Fig. 1 The closed five-compartment model of aquatic ecology

$$\begin{aligned} \frac{dq_1}{dt} &= - (k_{12} + k_{13} + k_{14} + k_{15}) q_1 + k_{21} q_2 + k_{31} q_3 + k_{51} q_5 \\ \frac{dq_2}{dt} &= k_{12} q_1 + k_{42} q_4 - (k_{21} + k_{24}) q_2 \\ \frac{dq_3}{dt} &= k_{13} q_1 - k_{31} q_3 \\ \frac{dq_4}{dt} &= k_{14} q_1 + k_{24} q_2 - k_{42} q_4 \\ \frac{dq_5}{dt} &= k_{15} q_1 - k_{51} q_5 \end{aligned}$$

由试验数据和初始条件, 经计算机拟合, 得水生生态系中水 (C_1)、底泥 (C_2)、鱼 (C_3)、螺蛳 (C_4)、水葫芦 (C_5) ⁹⁵Zr 浓度 C 随时间 t 变化的数学模式如下：

$$\begin{aligned} C_1 &= 654.3447e^{-2.2147t} + 0.3285e^{-0.006690t} + 0.1119e^{-0.0129t} + 0.1099e^{-0.1848t} + 0.0006e^{-0.3378t} \\ C_2 &= 85.7971e^{-2.2147t} + 1.6024e^{-0.006690t} + 91.8122e^{-0.0129t} + 2.9428e^{-0.1848t} + 0.3845e^{-0.3378t} \\ C_3 &= (18995.7705e^{-2.2147t} + 119516.3543e^{-0.006690t} + 682.5725e^{-0.0129t} + 656.1561e^{-0.1848t} + 5.3418e^{-0.3378t}) / m_3 \cdot 0 \\ C_4 &= (0198.6263e^{-2.2147t} + 304.2514e^{-0.006690t} + 679.3532e^{-0.0129t} + 155.6352e^{-0.1848t} + 9201.0370e^{-0.3378t}) / m_4 \cdot 0 \\ C_5 &= (13794.5841e^{-2.2147t} + 4717.3065e^{-0.006690t} + 6210.0565e^{-0.0129t} + 212256.7591e^{-0.1848t} + 23.4206e^{-0.3378t}) / m_5 \cdot 0 \end{aligned}$$

相关系数, 水 $r = 0.92$, 底泥 $r = 0.91$, 鱼 $r = 0.69$, 螺蛳 $r = 0.97$, 水葫芦 $r = 0.96$ 。其中 $m_x \cdot 0$ 为 t 时刻 x 分室的质量。转移系数 (l^{-1}) 分别为 $k_{12} = 0.5765$ $k_{13} = 0.0506$ $k_{14} = 0.0024$ $k_{15} = 0.0246$ $k_{21} = 0.0195$ $k_{31} =$

0.0152 $\chi_{51}=0.0169$ $\chi_{24}=0.0001$ $\chi_{42}=0.3241$ 。

2.5 水生生物对⁹⁵Zr 的富集系数 CF

水生生物对水体中放射性核素的富集系数定义为 水生系统的某一组分中放射性核素的比活度与同一时刻空间点上水中该核素的比活度之比,通常以 CF 表示。水生生物对水中⁹⁵Zr 浓集系数 CF 与时间关系列于表 4。

表 4 不同时期 ⁹⁵ Zr 在水生生物水葫芦、螺蛳、鱼中的富集系数值										
Table 4 Change in CF values in aquatic organisms in water hyacinth, snail and whole fish										
时间 Time (d)		1	2	4	6	9	15	22	29	34
富集系数	水葫芦 water hyacinth	12.67	18.09	25.37	36.69	31.22	41.75	165.59	295.26	396.57
CF	螺蛳 snail	1.51	1.43	1.19	1.24	0.89	1.67	6.53	11.90	15.83
	全鱼 Whole fish	0.35	0.94	0.41	0.06	0.32	0.33	0.34	0.45	0.30

水葫芦对水体中的⁹⁵Zr 富集作用最强,其次是螺蛳,鱼最小。其 CF 值随时间逐渐增大,至试验结束时(4d),水葫芦高达 396.57,螺蛳为 15.83。鲫鱼的主要可食部位(鱼肉)对⁹⁵Zr 的 CF 值几乎均为 0,表明鲫鱼对水中的⁹⁵Zr 不具有富集作用。锆不是生命必须元素,具有普遍的不溶性和非常低的生物吸收,所以它们的同位素⁹⁵Zr 显示了很低的食物流动性。

3 结论

(一) 从水中引入的⁹⁵Zr 由于沉降、土壤吸附、被水葫芦、螺蛳和鲫鱼吸附、吸收、摄入而致使水中的⁹⁵Zr 比活度呈快速下降。1d 后,已下降至初始比活度的 27.11%。

(二) 在水生态系中,不同的生物对⁹⁵Zr 的富集能力有很大差异。水葫芦对⁹⁵Zr 的富集能力远远大于螺蛳和鲫鱼对⁹⁵Zr 的富集能力,而螺蛳对⁹⁵Zr 的富集能力大于鲫鱼对⁹⁵Zr 的富集能力。鲫鱼对水中的⁹⁵Zr 不具有富集作用。

(三) 同一生物的不同部位对⁹⁵Zr 的富集能力也有差异,并且差异很大。

(四) 进入淡水生态系统中的⁹⁵Zr 在系统各组分中的动态变化规律可用示踪动力学分室模型描述 在本研究条件下,其定量表达式达到显著甚至极显著水平。

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