

环境科学

(HUANJING KEXUE)

ENVIRONMENTAL SCIENCE

第35卷 第7期

Vol.35 No.7

2014

中国科学院生态环境研究中心 主办

科学出版社 出版



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《环境科学》征稿简则(2679) 《环境科学》征订启事(2735) 信息(2476,2556,2713,2770)

三峡水库消落区土壤、植物汞释放及其在斑马鱼体的富集特征

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摘要: 利用室内模拟试验, 探讨了三峡水库消落区淹没后土壤、植物汞释放特征及其在斑马鱼体的富集水平. 结果表明, 随淹水时间的延长, 淹没土壤中总汞 (THg) 含量下降; 水体 THg 浓度总体明显升高. 淹没土壤及水体中甲基汞 (MMHg) 含量总体明显上升, 其中稗草 + 土壤处理水体中增加尤为明显, 淹水 21 d 后, 其浓度是土壤处理的 2.52 倍. 表明消落区土壤、植物是水库水体汞的一个重要来源. 供试稗草淹水分解造成水体 pH 及溶解氧 (DO) 含量下降、溶解性有机碳 (DOC) 含量上升, 对土壤 MMHg 含量无明显影响, 对水体 MMHg 影响较大. 供试斑马鱼头部、内脏及肌肉中 THg 含量总体明显上升, 与水体中 THg 浓度具有显著相关性 ($P < 0.01$). 鱼体头部、内脏及肌肉中均出现不同程度的 MMHg 富集现象, 以头部与肌肉最为明显. 淹水 21 d 后, 添加土壤处理的鱼体头部、内脏及肌肉 MMHg 增加的含量分别是对照的 1.75 ~ 6.25、3.53 ~ 8.38、2.22 ~ 3.36 倍; 稗草 + 土壤处理的分别是土壤处理的 3.57、2.37、1.52 倍. 可见, 淹没土壤是鱼体 MMHg 增加的重要来源, 淹没植物改变原水环境条件影响其向水体释汞过程能提高鱼体中 MMHg 含量.

关键词: 消落区; 土壤; 植物; 汞; 斑马鱼

中图分类号: X131; X173; X174 文献标识码: A 文章编号: 0250-3301(2014)07-2721-07 DOI: 10.13227/j.hjxx.2014.07.040

Release of Mercury from Soil and Plant in Water-Level-Fluctuating Zone of the Three Gorges Reservoir Area and Its Accumulation in Zebrafish

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Abstract: To investigate the production, distribution and bioavailability of methylmercury (MMHg) in soil and plants of the water-level-fluctuating zone (WLFZ) of the Three Gorges Reservoir area, simulation experiments were conducted in laboratory. Results indicated that the level of total mercury (THg) in soil decreased with the lengthening of submerging time while that in water increased obviously. The level of MMHg in inundated soil and water increased, especially in the water treated by *Echinochloa crusgalli* and soils. And the MMHg level in that treatment was 2.52 times higher than that treated only by soils for 21 days. This indicated that soil and plants of WLFZ were important sources of mercury in the water of the reservoir. *Echinochloa crusgalli* as the tested plant was decomposed after being submerged, leading to lower pH and DO and higher DOC, which had little effect on MMHg in soil but significant effect on MMHg in water. The level of THg in the head, viscera and muscle of zebrafish increased obviously, which had a significant correlation with that in water ($P < 0.01$). MMHg levels accumulated in the head, viscera and muscle of zebrafish differed to some degree, particularly in the head and muscle. After treated in the soils for 21 days, MMHg levels in the head, viscera and muscle of zebrafish were 1.75-6.25, 3.53-8.38 and 2.22-3.36 times higher than those in the control groups, respectively. While for the treatment of *Echinochloa crusgalli* and soil, MMHg levels in zebrafish's head, viscera and muscle were 3.57, 2.37 and 1.52 times higher than those treated only by soil, respectively. Therefore, submerged soil was the main source of MMHg in fish. And submerged plants changed the water condition and affected the release of mercury to water so as to cause elevated levels of MMHg in fish.

Key words: water-level-fluctuating zone (WLFZ); soil; plant; methylmercury; zebrafish

收稿日期: 2013-12-16; 修订日期: 2014-02-28

基金项目: 国家重点基础研究发展规划(973)项目(2013CB430004); 国家自然科学基金项目(41373113, 41173116); 中国博士后科学基金项目(2013M542242)

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壤^[11]及鱼体^[12]中 MMHg 含量, DMA-80 固体进样自动测汞仪直接测定土壤及鱼样中 THg 含量. DOC 的测定: 取水样过 0.45 μm 后, 用全自动 TOC 分析仪 (Sievers InnovOx, GE, USA) 测定. 试验期间定期测定水体中 pH (Sartorius PT-10, German)、溶解氧 DO (YSI550A, USA).

1.4 质量控制及数据处理

试验所使用的玻璃器皿在使用前均用硝酸 (25%, 体积比) 浸泡 24 h 以上, 超纯水清洗后经马弗炉 500℃ 灼烧 30 min, 在洁净无汞的环境下冷却后使用. 分析过程中采用空白试验、标准物质测定、平行样控制及加标回收率进行质量控制. 土样标准物质为 GBW07405 (GSS-5), 鱼样标准参考物质为干鱼样 NRCC-TORT-2. 本试验对于固体样品、水样总汞测定加标回收率分别为 93% ~ 112%、

88% ~ 109%; MMHg 测定加标回收率则分别为 86% ~ 109%、89% ~ 113%.

数据分析与作图分别使用软件 SPSS 16.0 与 Origin 8.1.

2 结果与讨论

2.1 水环境参数的变化

由表 3 可见, 淹水后, 处理 A 中 pH 无明显变化. 处理 B 和 C 中 pH 在淹水初期明显下降, 且低于处理 A, 与土壤 pH 趋于一致, 7 d 后测得的 pH 值变化不大. 水体中 DO 含量均随时间的延长而降低, 呈处理 C < B < A. 与处理 A 相比, 处理 B 和 C 水体中溶解性有机碳 (DOC) 均明显升高, 且处理 C 的 DOC 远高于 B、A, 说明土壤淹没后会向水体释放有机质, 而植物的腐解则是水体 DOC 的重要来源^[13].

表 3 试验期间水体中 pH 值、DO、DOC 浓度变化

Table 3 Variations of pH, DO and DOC in the water during the experiment

时间/d	pH			DO/mg·L ⁻¹			DOC/mg·L ⁻¹		
	A	B	C	A	B	C	A	B	C
1	8.45	8.50	8.46	6.98	7.06	7.11	3.44	3.56	3.43
7	8.43	8.01	7.88	6.54	6.23	5.50	3.19	4.58	9.50
14	8.34	7.98	7.82	5.87	5.74	4.45	2.98	4.67	8.85
21	8.35	8.19	7.80	5.43	4.83	3.09	3.23	3.87	8.56

2.2 总汞的释放特征

由图 1(a) 可见, 淹水后处理 B 与 C 土壤中总汞 (THg) 含量均降低, 说明土壤淹水后, 土壤汞会向水体释放. 但淹水 7 d 后, 系统中土壤 THg 含量变化不明显, 基本达到平衡. 土壤被淹没后所释放的汞只占其 THg 量的一小部分^[14], 本研究淹水 21 d 后, 处理 B

与 C 土壤 THg 含量分别下降了 18.49%、15.62%.

处理 A 水体中 THg 浓度随淹水时间延长而逐渐下降, 而处理 B 与 C 由于有土壤、植物汞释放的补充, 水体 THg 浓度总体明显升高, 并呈先增后减的趋势 [图 1(b)], 其中在第 7 d 测得处理 B、C 水体 THg 浓度最高, 分别为 17.90 ng·L⁻¹、25.78

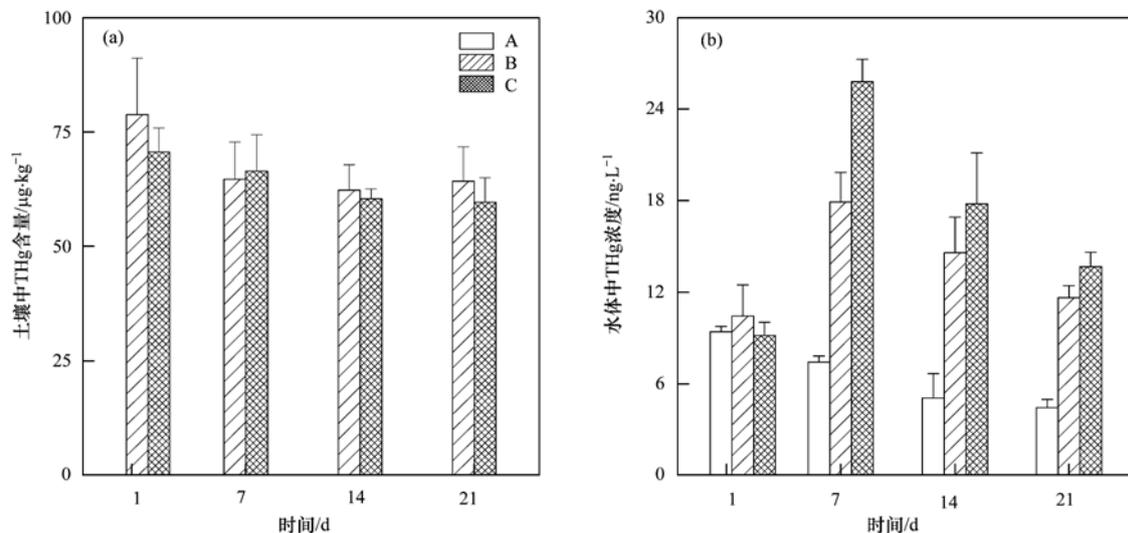


图 1 土壤及水体中 THg 的变化趋势

Fig. 1 Variations of THg levels in soil and water

$\text{ng}\cdot\text{L}^{-1}$,之后各处理 THg 浓度均降低,这可能与 THg 向大气释放、被鱼体富集及土壤再吸附有关.由图 1(b)还可看出,处理 B、C 水体 THg 浓度远高于对照(处理 A),而添加植物的处理 C 水体 THg 浓度明显高于处理 B,这一现象进一步证明了土壤、植物淹水后能迅速释放 THg^[15,16].在淹水试验后的第 21 d,处理 B 与 C 的水体 THg 浓度分别是 A 的 2.62、3.07 倍,表明三峡水库消落区土壤、植物是库区水体 THg 的一个重要来源.

2.3 甲基汞的释放特征

由图 2(a)可见,处理 B 与 C 中土壤 MMHg 含

量随时间变化均明显升高,在淹水的一周内增加较为明显,这是由于淹水厌氧环境有利于汞的微生物甲基化,进而提高土壤中 MMHg 含量^[17].在淹水 21 d 后,处理 B 与 C 的 MMHg 含量分别为 $956.63 \text{ ng}\cdot\text{kg}^{-1}$ 、 $1139.79 \text{ ng}\cdot\text{kg}^{-1}$,是第 1 d 的 2.20、2.69 倍,表明了库区消落区土壤淹水后利于土壤中 MMHg 的产生.淹水初期,处理 B 与 C 土壤中 MMHg 含量无明显差异($P>0.05$),但试验 14 d 后,添加植物的处理 C 土壤 MMHg 含量明显高于处理 B,说明随着时间的延长,淹没植物分解释放的 DOC、营养元素等对土壤 MMHg 的产生有影响.

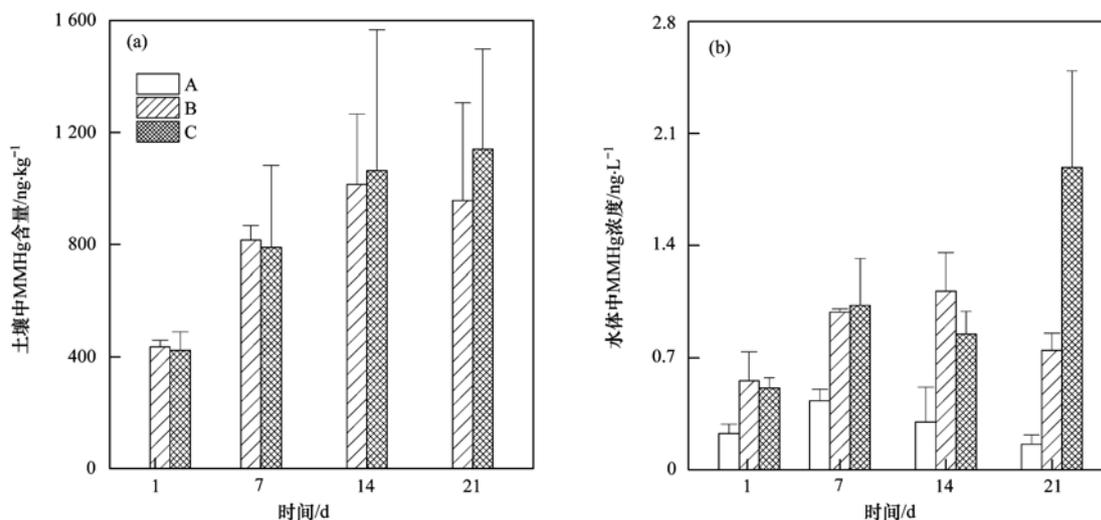


图 2 土壤与水体 MMHg 的含量变化

Fig. 2 Variations of MMHg in soil and water

水体中 MMHg 变化见图 2(b),与对照相比,处理 B 与 C 中 MMHg 浓度总体均明显上升.其中,处理 B 中 MMHg 浓度呈先增后减的趋势,在第 14 d 测定值最高,为 $1.12 \text{ ng}\cdot\text{L}^{-1}$,随时间延长,MMHg 浓度下降,可能与水体去甲基化作用、鱼体富集及土壤再吸附等有关.处理 C 与 B 水体中 MMHg 浓度在前 14 d 无明显差异($P>0.05$),但在淹水 21 d 后,达 $1.89 \text{ ng}\cdot\text{L}^{-1}$,是 B 的 2.52 倍.这是由于随着时间的延长,处理 C 添加的植物腐解使水体中 DOC 含量上升、DO 下降(表 3),利于汞的甲基化,从而提高水体中 MMHg 浓度.

淹水试验 21 d 后,处理 B 与 C 水体中 MMHg 浓度分别是 A 的 4.67、11.79 倍,处理 B 与 C 水体的 MMHg 浓度变化与土壤中 MMHg 含量变化呈极显著相关($P<0.01$),表明了水中 MMHg 主要来源于土壤.此外,处理 B 与 C 土壤中 MMHg 占 THg 的百分比(MMHg: THg)呈明显上升趋势[图 3(a)],第

21 d 土壤中 MMHg: THg 分别是第 1 d 的 2.70、3.04 倍.有研究指出^[18],被淹没土壤中 MMHg 含量以及 MMHg 占 THg 的比例均会显著上升.处理 B 水体中 MMHg: THg 呈先降后升再降的趋势[图 3(b)],而 C 中在前 14 d 无明显差异,但第 21 d 时其 MMHg: THg 最高,达 13.84%,是 B 的 2.16 倍,说明随淹水时间延长,植物改变原水环境参数能一定程度提高水体中 MMHg: THg.

2.4 汞在鱼体中的富集特征

2.4.1 总汞在鱼体中的富集特征

试验 21 d 后,各处理鱼体平均体重为 $0.48 \text{ g} \pm 0.13 \text{ g}$,平均体长为 $4.20 \text{ cm} \pm 0.11 \text{ cm}$.随淹水时间延长,处理 A 头部、内脏和肌肉这 3 个部位 THg 含量基本稳定,但处理 B 与 C 总体呈明显上升趋势,并表现为 $C>B>A$ (图 4).21 d 时处理 B 头部、内脏及肌肉中 THg 含量分别比初始的增加 32.77%、40.56% 与 42.21%,而 C 的则增加了 49.09%、

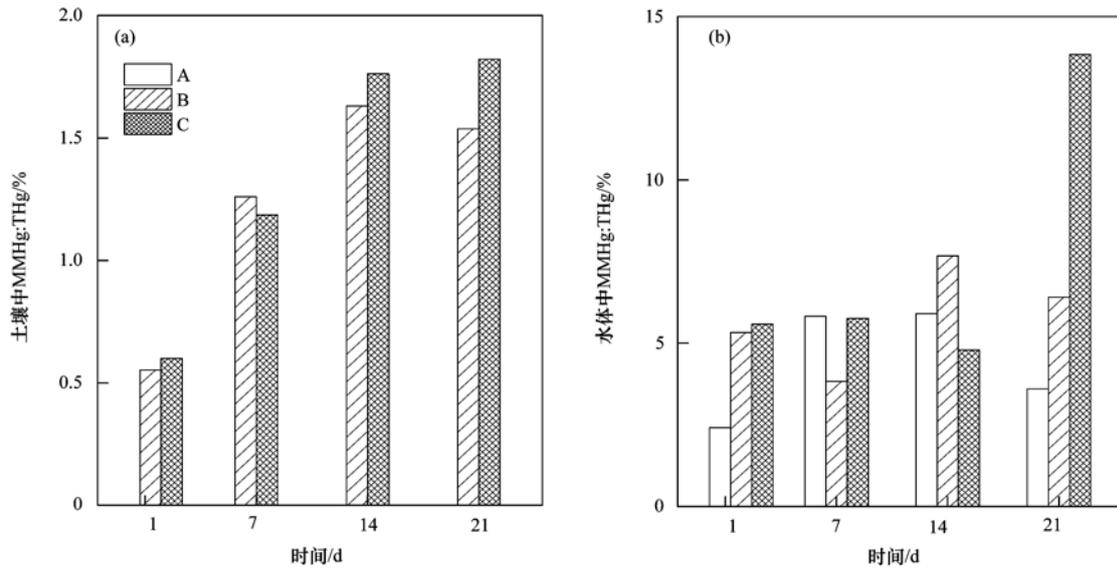


图3 土壤与水体 MMHg:THg 百分比变化

Fig. 3 Variations of the percentage of MMHg in THg in soil and water

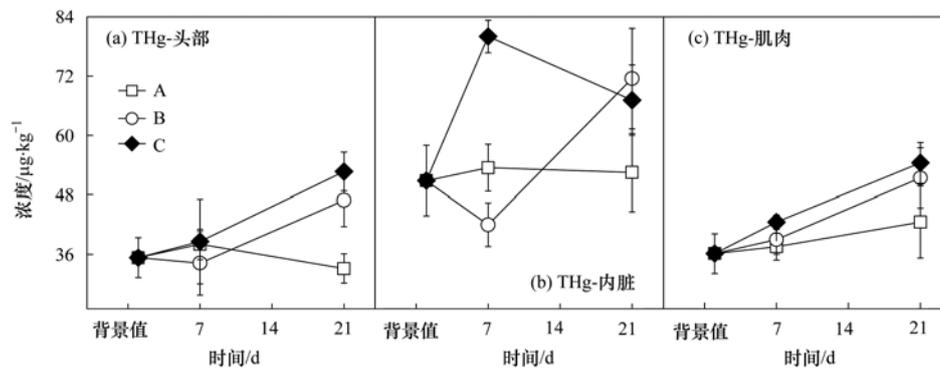


图4 斑马鱼体中的 THg 含量

Fig. 4 Variations of THg levels in zebrafish

31.98%与50.51%，且与土壤和水体中 THg 呈显著正相关 ($P < 0.01$)，表明淹没土壤与植物向水体释汞是鱼体 THg 增加的直接来源。

2.4.2 甲基汞在鱼体中的富集特征

由图 5 可见，随着时间的延长，各处理鱼体 MMHg 含量均呈上升趋势。第 21 d 时，各处理鱼体头部、内脏及肌肉中 MMHg 含量分别增加了 0.40~2.52、0.12~1.58、0.69~2.30 倍。可见，斑马鱼的 3 个部位均有 MMHg 富集效应，其中，头部与肌肉中 MMHg 的积累最为明显。以往的研究也表明，鱼体对 MMHg 富集有累积迅速、排除缓慢的特征^[19]。

各处理斑马鱼头部均呈现较明显的 MMHg 净累积，其原因可能是，MMHg 能通过非 Na^+ 依赖型中性氨基酸载体（系统 L）顺利进入血脑屏障^[20]，大量

累积在脑部^[21]。另一方面，腮部是水及离子交换的主要场所^[19]，可能存在无机汞的甲基化^[22]。本试验 21 d 后，处理 C 的 MMHg 积累速率比 B 中高，两者分别为 $1.33 \mu\text{g}\cdot(\text{kg}\cdot\text{d})^{-1}$ 、 $0.37 \mu\text{g}\cdot(\text{kg}\cdot\text{d})^{-1}$ ，且均高于处理 A。说明淹没植物及土壤有增加鱼体中 MMHg 含量的风险。

作为解毒及贮存器官，肝脏可能因参与到污染物代谢中而富集大量的污染物^[21]。在 21 d 时，处理 C 中的鱼体内脏富集 MMHg 最明显，分别增加 $22.52 \mu\text{g}\cdot\text{kg}^{-1}$ 。但整体上各处理内脏的 MMHg:THg 变化不大，这可能与肝脏中的解毒过程有关，肝脏能将 MMHg 去甲基化转化成 $\text{Hg}(\text{II})$ ^[23]，也能通过血液运输将 MMHg 贮存于肌肉中^[24]。

肌肉中甲基汞含量变化与头部类似，在第 7 d，处理 B 鱼体肌肉中 MMHg 呈明显的上升趋势，这与

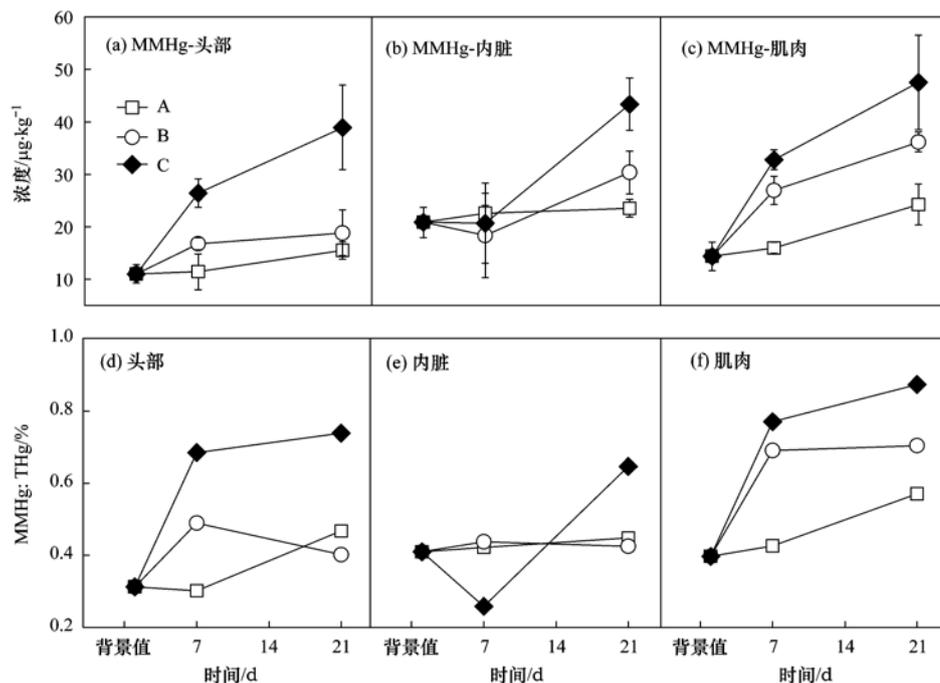


图5 斑马鱼体中MMHg含量及MMHg:THg的变化趋势

Fig. 5 Variations of MMHg levels and the percentage of MMHg in THg in zebrafish

水体中的MMHg变化趋势相吻合. 第21 d时,处理C肌肉中MMHg含量最高($47.50 \mu\text{g}\cdot\text{kg}^{-1}$). 与无机汞相比,MMHg能稳定穿过生物膜,积累在肌肉内^[2, 25, 26]. 且MMHg主要以半胱氨酸-硫醇复合物存在于肌肉中^[27],其半衰期为400 d左右^[28]. 本试验过程中未添加食物,处理B、C斑马鱼肌肉中MMHg含量与土壤、水体MMHg均呈显著正相关($P < 0.05$),表明斑马鱼体MMHg主要来自试验系统中土壤、植物释放.

通常鱼体富集MMHg有两种途径,一是从水体吸入,二是通过食物摄取. 研究发现,鱼体直接从水体吸入的MMHg量十分有限^[29],其MMHg主要来源于摄入食物^[30]. 处理A中鱼体的头部、内脏及肌肉三部分的MMHg含量都略有上升,说明了当没有食物来源时,从水体吸入会变成鱼体富集MMHg的主要途径. 对于同一水体,鱼体汞一般比水体汞高6个数量级^[31],水库被淹没后,水体中MMHg浓度的微弱增加可能会导致鱼体MMHg的显著升高^[14]. 试验21 d中,添加土壤处理的鱼体头部、内脏及肌肉MMHg增加的含量分别是对照的1.75~6.25、3.53~8.38、2.22~3.36倍;添加稗草处理的分别是未添加的3.57、2.37、1.52倍,进一步验证了淹没土壤和植物是水库鱼体MMHg增加的重要来源.

3 结论

三峡水库消落区淹没土壤、植物是水体汞升高的重要来源;淹没土壤与植物向水体释汞过程会明显增加鱼体中THg、MMHg含量;淹没植物的腐解,一方面可释放所含的汞,增加水体汞含量,同时,所释放的DOC等营养物质可促进汞的甲基化,提高生态风险.

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环 境 科 学

(HUANJING KEXUE)

(月刊 1976年8月创刊)

2014年7月15日 第35卷 第7期

ENVIRONMENTAL SCIENCE

(Monthly Started in 1976)

Vol. 35 No. 7 Jul. 15, 2014

主 管	中国科学院	Superintended	by	Chinese Academy of Sciences
主 办	中国科学院生态环境研究中心	Sponsored	by	Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences
协 办	(以参加先后为序) 北京市环境保护科学研究院 清华大学环境学院	Co-Sponsored	by	Beijing Municipal Research Institute of Environmental Protection School of Environment, Tsinghua University
主 编	欧阳自远	Editor-in -Chief		OUYANG Zi-yuan
编 辑	《环境科学》编辑委员会 北京市2871信箱(海淀区双清路 18号, 邮政编码:100085) 电话:010-62941102, 010-62849343 传真:010-62849343 E-mail: hjkx@rcees. ac. cn http://www. hjkx. ac. cn	Edited	by	The Editorial Board of Environmental Science (HUANJING KEXUE) P. O. Box 2871, Beijing 100085, China Tel:010-62941102, 010-62849343; Fax:010-62849343 E-mail: hjkx@rcees. ac. cn http://www. hjkx. ac. cn
出 版	科 学 出 版 社 北京东黄城根北街16号 邮政编码:100717	Published	by	Science Press 16 Donghuangchenggen North Street, Beijing 100717, China
印 刷 装 订	北京北林印刷厂	Printed	by	Beijing Bei Lin Printing House
发 行	科 学 出 版 社 电话:010-64017032 E-mail: journal@ mail. sciencep. com	Distributed	by	Science Press Tel:010-64017032 E-mail: journal@ mail. sciencep. com
订 购 处	全国各地邮电局	Domestic		All Local Post Offices in China
国外总发行	中国国际图书贸易总公司 (北京399信箱)	Foreign		China International Book Trading Corporation (Guoji Shudian), P. O. Box 399, Beijing 100044, China

中国标准刊号: ISSN 0250-3301
CN 11-1895/X

国内邮发代号: 2-821

国内定价: 90.00元

国外发行代号: M 205

国内外公开发行