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 CHEN Zong-ya, WANG Yong-jie, SHU Rui, et al. Effects of straw amendment on methylmercury accumulation in soil and crop plants under wheat–rice rotation[J]. *Journal of Agro-Environment Science*, 2016, 35(10): 1931–1936.

秸秆覆盖还田对稻麦轮作体系中土壤及作物甲基汞累积的影响

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摘要:以贵州万山汞矿区的汞污染稻田土为研究对象,采用盆栽模拟探究了小麦-水稻轮作情况下秸秆覆盖还田对土壤及小麦与水稻在重要生育期(出苗期、分蘖期、拔节期、扬花-灌浆期、乳熟-收获期)甲基汞累积的影响。结果表明:秸秆还田能提高土壤中甲基汞含量,且不同作物秸秆还田对土壤甲基汞及作物甲基汞的影响存在显著差别,其中,小麦秸秆还田后土壤甲基汞含量增加127.1%,水稻秸秆还田后土壤甲基汞含量增加25.1%,这可能缘于迥异的种植条件。同时,作物秸秆还田后小麦、水稻植株体内甲基汞含量也呈增加趋势,其中,小麦根部、地上部(茎叶)和籽粒甲基汞含量分别增加124.6%、79.2%和169%,水稻根部、地上部和籽粒甲基汞含量依次增加40.1%、61.7%和25.9%,表明作物甲基汞吸收累积分量的上升可能主要源于土壤甲基汞含量的上升。因此,汞污染地区传统农艺措施中惯常采用的秸秆还田可能会显著增加人体甲基汞暴露风险,在汞污染地区推广作物秸秆还田措施时应持谨慎态度。

关键词:秸秆还田;稻田土壤;稻麦轮作;作物;甲基汞

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Effects of straw amendment on methylmercury accumulation in soil and crop plants under wheat–rice rotation

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Abstract: There is growing concern about methylmercury(MeHg) accumulation in crop plants in Hg-contaminated areas. To investigate the possible influences of straw amendment on soil methylmercury levels and its accumulation in crop plants, mercury-contaminated paddy soil was collected from Wanshan mercury mine area and amended with 1% (W/W) crop straw(wheat or rice) in a wheat–rice rotation system. At different stages(e.g., seedling, tillering, jointing, flowering and harvesting) of plant growth, the soil and plant samples were collected and analyzed for MeHg. The results show that straw amendment could lead to a substantial increases in MeHg levels in both soils and plants. Soil MeHg increased by 127.1% under wheat-straw amendment and 25.1% under rice-straw amendment, respectively. This was possibly due to the different planting conditions. Correspondingly, MeHg levels in root, aboveground part and grain of wheat increased by 124.6%, 79.2% and 169%, respectively, compared to the control. Similarly, MeHg levels in root, aboveground part and grain of rice increased by 40.1%, 61.7% and 25.9%, respectively. Overall, there is a positive relationship between the crop plant MeHg levels and soil MeHg levels under wheat–rice rotation, indicating that straw amendment could increase soil MeHg levels and subsequently the MeHg accumulation in crops,

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which could enhance the potential risk of human exposure to MeHg in mercury-contaminated areas. These results suggest that it should be cautious when promoting straw amendment in mercury-contaminated areas.

Keywords: straw return; paddy soil; wheat-rice rotation; crop; methylmercury

2014年全国土壤污染状况调查公报显示,我国土壤汞污染点位超标率为1.6%^[1],尤其是在工矿业废弃地,土壤汞污染较为突出^[2-3]。贵州万山地区作为我国最大的汞矿区,虽早已停止开采,但该矿区的土壤汞含量仍高达790 mg·kg⁻¹^[4]。近期研究表明,稻田能促使土壤中汞的甲基化,虽然其占总汞的比例通常小于1%^[5],但高毒性的甲基汞易在稻米中富集^[6],导致汞矿区稻米甲基汞含量可达145 μg·kg⁻¹^[2],这对当地居民的身体健康构成了极大威胁^[7-8]。因此,稻米的摄入已成为我国内陆居民甲基汞暴露的重要途径之一^[8-10]。通常,作物收获后其秸秆会被提倡以还田的方式加以利用,我国每年还田的作物秸秆超过1.5亿t,且仍在快速增加^[11]。关于作物秸秆还田对土壤及农作物品质的影响,近年来的研究大多集中在作物秸秆还田可增加土壤中有机质含量^[12-13]、为植物生长提供营养、使作物增产等^[14-15]方面;而在秸秆还田对土壤中汞的生物地球化学行为影响方面仅有少量报道,如有研究表明,淹水条件下添加水稻秸秆能促使土壤中甲基汞含量升高^[16-18],但稻米中甲基汞含量增加不明显^[16],而在稻麦轮作条件下作物秸秆还田对土壤汞净化甲基化及作物甲基汞累积会造成何种影响,目前尚无相关研究。为明确秸秆还田对稻麦轮作体系中土壤及不同作物甲基汞累积的影响,有必要针对汞矿区秸秆还田可能带来的汞污染暴露风险开展深入的研究工作。本文采集万山汞矿区的汞污染稻田土开展盆栽试验,模拟秸秆覆盖还田方式下水稻-小麦轮作情况时,作物生长的重要时期(出苗期、分蘖期、拔节期、扬花-灌浆期、乳熟-收获期)植株和土壤样品中甲基汞含量变化特征,旨在探索秸秆还田对汞污染稻田土壤汞转化及作物甲基汞累积的影响,为汞污染地区制定合理的作物秸秆还田措施提供科学依据。

1 材料与方法

1.1 实验材料

供试土壤样品:采自贵州省万山汞矿区五坑(27°32'N, 109°14'E)的稻田土壤,经室内风干,研磨,过2 mm筛备用。基本理化参数:全氮0.18%,全碳2.89%,总有机碳2.49%,pH值8.04,土壤粘粒36.6%,粗粉砂18.9%,总汞含量(58±3) mg·kg⁻¹,甲基汞含量(1.62±

0.41) μg·kg⁻¹。

供试作物秸秆:水稻、小麦秸秆均采自未受汞污染的贵州省贵阳市花溪区。秸秆洗净,30℃烘干至恒重,磨碎过2 mm筛备用。水稻秸秆总汞含量0.33 mg·kg⁻¹,甲基汞含量0.083 μg·kg⁻¹;小麦秸秆总汞含量0.19 mg·kg⁻¹,甲基汞含量0.061 μg·kg⁻¹。供试作物品种:烟农19号小麦,中优808水稻。

1.2 实验设计

供试土壤按总氮100 mg·kg⁻¹、总磷100 mg·kg⁻¹、总钾143 mg·kg⁻¹施加底肥后混匀,根据表1实验方案开展小麦种植(12月到次年5月),第二茬淹水种植水稻(次年6月到11月),种植水稻前土壤经风干,再次过2 mm筛以去除残留麦根的影响。具体方法:每盆3.5 kg土壤+35 g秸秆(即土壤和秸秆质量比为100:1)^[19-20],加静置数日的自来水平衡2 d,撒播饱满的小麦种子8~10粒;水稻季淹水10 d后,插播20 d的优质水稻秧7~8株(无汞污染土壤培育秧苗)。所有盆栽实验均在室外进行,但采用玻璃板防止雨水的影响。作物生长期,小麦种植土壤含水率在13.5%~17.6%之间,水稻种植土壤淹水层在3~6 cm。同时,进行例行的虫害、肥料管理。

作物盆栽种植周期为小麦140 d、水稻120 d,均选取重要的5个时期:出苗期、分蘖期、拔节期、扬花-灌浆期、乳熟-收获期,连根采集植株2株,同时采集根区土壤。植株用去离子水洗净后,用0.8 mmol·L⁻¹半胱氨酸溶液浸泡10 min^[21],以去除植株表面吸附的甲基汞。根部去除铁膜^[22],去离子水清洗3遍,冻干、粉碎,密封保存待测;土壤样品冻干、研磨过100目筛,密封保存待测。

1.3 测定方法

土壤与作物样品甲基汞测定,均采用25% KOH-CH₃OH方法消解^[23],利用自动甲基汞仪(Model III,

表1 实验设计方案

Table 1 Designed of pot experiments

| 项目 | 小麦种植季 | | 水稻种植季 | | 平行样 |
|-----|-----------------|------|-----------------|------|-----|
| | 还田秸秆 | 播种方式 | 还田秸秆 | 播种方式 | |
| 对照组 | 无 | 种子 | 无 | 幼苗 | 6 |
| 还田组 | 1%水稻秸秆 (W/W) | 种子 | 1%小麦秸秆 (W/W) | 幼苗 | 6 |

Brooks,美国,参考美国EPA方法1630)进行测定分析。选用河口沉积物(ERM-CC580,Belgium)和鱼蛋白(DORM-3,Canada)标准物质,同时采用加标回收方法,以检验实验分析的准确性,所得回收率在94%~102%。

1.4 统计分析方法

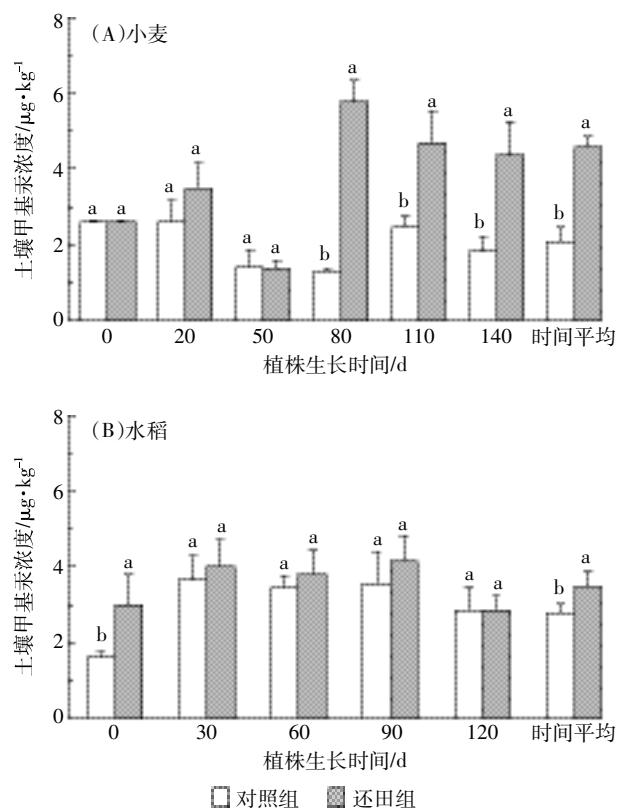
土壤与作物甲基汞含量变化均采用单因素方差分析(One-way ANOVA)进行统计学检验。检验组间差异使用两两比较(Tukey' HSD),并用Origin 8.5进行绘图。

2 结果与讨论

2.1 秸秆还田对稻田土壤甲基汞浓度的影响

秸秆还田增加了土壤中甲基汞含量(图1),随着小麦生长时间的变化,土壤中甲基汞含量呈现一个动态变化趋势(图1A)。秸秆还田组土壤甲基汞含量在1.36~5.78 $\mu\text{g}\cdot\text{kg}^{-1}$ 间波动;对照组的土壤甲基汞含量在1.42~2.45 $\mu\text{g}\cdot\text{kg}^{-1}$ 间波动。秸秆还田组土壤甲基汞平均含量($4.59 \mu\text{g}\cdot\text{kg}^{-1}$)是对照组甲基汞含量($2.07 \mu\text{g}\cdot\text{kg}^{-1}$)的2.2倍。秸秆还田后的50 d内(拔节期前),土壤中甲基汞含量变化不大(平均含量为 $2.42\pm0.59 \mu\text{g}\cdot\text{kg}^{-1}$);秸秆还田50 d后土壤甲基汞含量迅速增加(平均为 $5.04\pm0.48 \mu\text{g}\cdot\text{kg}^{-1}$),是50 d前的2.1倍。可见,水稻秸秆还田对土壤汞净甲基化是一个持续影响过程,可能原因是前期(12月到次年3月)气温较低导致土壤微生物活性较低造成秸秆分解缓慢,后期(次年3月到5月)随着气温回升,土壤中微生物活性增强促使秸秆分解后产生大量的活性有机碳^[5,24],在微生物与活性有机碳的共同作用下促进汞的净甲基化,最终导致土壤中甲基汞含量增多,使其与对照组的差异日趋明显。

图1B显示,添加小麦秸秆淹水10 d(图中0 d)后,秸秆还田组土壤甲基汞含量($2.99 \mu\text{g}\cdot\text{kg}^{-1}$)是对照组土壤甲基汞含量($1.59 \mu\text{g}\cdot\text{kg}^{-1}$)的1.9倍。但淹水40 d后(图中30 d,拔节期),对照组的土壤甲基汞含量在($3.37\pm0.38 \mu\text{g}\cdot\text{kg}^{-1}$)上下波动,秸秆还田组土壤甲基汞含量在($3.70\pm0.59 \mu\text{g}\cdot\text{kg}^{-1}$)上下波动,略高于对照组,但无显著性差异。小麦秸秆还田初期土壤中汞的净甲基化加快可能与厌氧微生物参与下小麦秸秆快速分解有关^[25]。有研究表明,淹水36 d内小麦秸秆以 $64.9 \text{ mg}\cdot\text{d}^{-1}$ 的腐解速率快速分解^[26],分解产生的溶解性有机质(DOM)和硫酸盐还原产生的还原性硫(S)可能与无机汞络合形成可被汞甲基化细菌所利用



图中不同字母代表各处理组间差异显著($P<0.05$)。下同
Different letters indicate significant differences, $P<0.05$. The same below.

图1 秸秆还田后土壤甲基汞浓度变化

Figure 1 Concentrations of MeHg in soil with wheat planting and rice planting after straw amendment during plant growth period

的Hg-S-DOM三元络合物^[27-28],促使无机汞转化为毒性更强的甲基汞。随着淹水时间的增长,厌氧条件趋于稳定,同时水稻根系发育分泌大量活性有机质^[29-30],导致秸秆还田对汞净甲基化的促进作用越来越不明显。总体来说,无论是在小麦种植时期还是在水稻种植时期,秸秆还田均能促使土壤汞的净甲基化而使土壤甲基汞含量增加。

2.2 秸秆还田对作物根部甲基汞浓度的影响

秸秆还田对作物根部甲基汞累积具有一定影响,根部甲基汞含量的变化总体与土壤甲基汞含量变化规律一致(图2)。在小麦种植后50 d内(图2A),虽然对照组小麦根部甲基汞累积量($4.19\sim8.60 \mu\text{g}\cdot\text{kg}^{-1}$)远高于秸秆还田组($2.46\sim4.52 \mu\text{g}\cdot\text{kg}^{-1}$)。但是小麦种植50 d以后,秸秆还田组小麦根部甲基汞的累积量逐渐增多,收获时达到 $22.29 \mu\text{g}\cdot\text{kg}^{-1}$,对照组小麦根部甲基汞的累积量变化不大,收获时为 $3.99 \mu\text{g}\cdot\text{kg}^{-1}$,仅为秸秆还田组的18%。

相关分析表明,水稻秸秆还田组小麦根部甲基汞

含量与土壤甲基汞含量有较好的正相关关系($r^2=0.695$, $P<0.05$), 说明根部甲基汞含量主要受土壤甲基汞含量的影响。水稻种植季(图2B, 生长30 d左右, 拔节期), 稻秆还田促进水稻根部甲基汞含量大幅增加, 其含量($32.95 \mu\text{g}\cdot\text{kg}^{-1}$)是对照组根部甲基汞含量($11.99 \mu\text{g}\cdot\text{kg}^{-1}$)的2.8倍; 水稻种植30 d以后, 稻秆还田组水稻根部甲基汞含量有所降低, 但与对照组没有显著性差异, 且其含量变化处于一个相对稳定的水平。另外, 就对照组小麦与水稻根部甲基汞平均含量(汞暴露期)而言, 小麦根部甲基汞含量($4.18 \mu\text{g}\cdot\text{kg}^{-1}$)仅为水稻根部甲基汞含量($13.92 \mu\text{g}\cdot\text{kg}^{-1}$)的 $\frac{1}{3}$, 说明在汞污染地区, 水稻根部对土壤中甲基汞有高累积性。总体上, 作物根部甲基汞累积量主要受土壤中甲基汞含量变化的影响。这一规律与已有研究结果一致^[31]。

2.3 稻秆还田对作物地上部甲基汞浓度的影响

稻秆还田在一定程度上能促进水稻与小麦植株地上部(均为茎叶混合部分, 不包括籽粒部分)对甲基汞的累积(图3)。苗期20 d时稻秆还田组小麦地上部甲基汞含量为 $1.78 \mu\text{g}\cdot\text{kg}^{-1}$, 50 d时甲基汞含量为

$0.97 \mu\text{g}\cdot\text{kg}^{-1}$, 与对照相比, 同时间点分别增加24.5%和59.4%。在收获期, 稻秆还田组小麦地上部甲基汞含量($4.09 \mu\text{g}\cdot\text{kg}^{-1}$)是对照组甲基汞($2.26 \mu\text{g}\cdot\text{kg}^{-1}$)的1.8倍(图3A)。显然, 水稻稻秆还田组小麦地上部甲基汞的含量明显高于对照组。

如图3B所示, 在水稻种植过程中, 植株生长30 d时, 稻秆还田组水稻地上部甲基汞含量($20.2 \mu\text{g}\cdot\text{kg}^{-1}$)是对照组的水稻地上部甲基汞含量($6.99 \mu\text{g}\cdot\text{kg}^{-1}$)的2.9倍。从30 d后到收获时, 稻秆还田组地上部甲基汞含量($4.77 \mu\text{g}\cdot\text{kg}^{-1}$)与对照组地上部甲基汞含量($4.92 \mu\text{g}\cdot\text{kg}^{-1}$)无明显差异。

上述结果表明, 在淹水条件下, 添加小麦稻秆能促使水稻植株地上部对甲基汞的累积, 主要集中在水稻植株生长的扬花期(60 d前)。另外, 就对照组小麦植株与水稻植株地上部甲基汞平均含量而言, 水稻植株地上部甲基汞含量是小麦植株甲基汞含量的4倍。值得注意的是, 小麦地上部甲基汞含量不断累积, 而水稻地上部甲基汞含量在扬花期后不断减少。Meng等^[32]研究表明, 在水稻扬花-灌浆期后, 大部分甲基汞

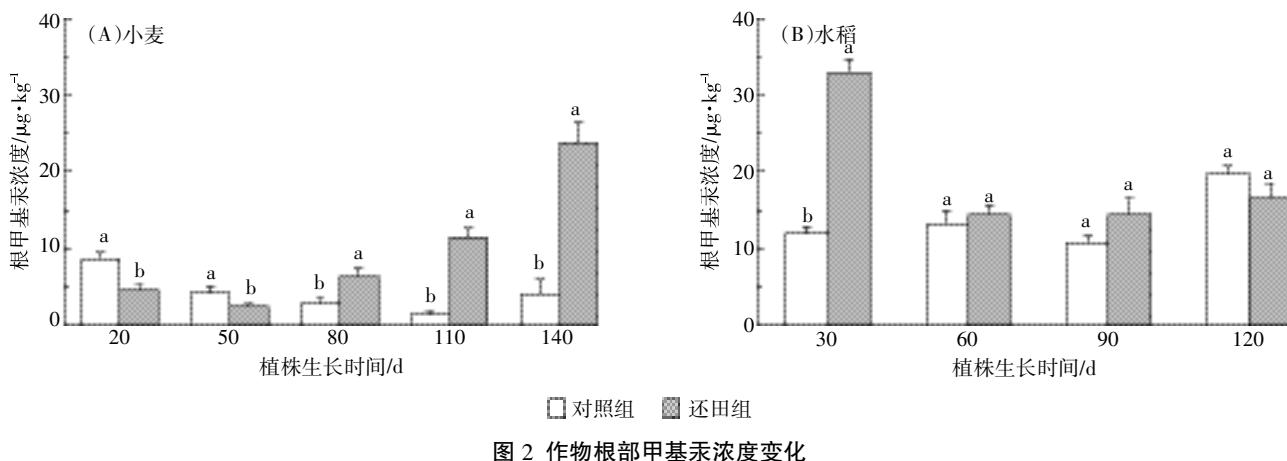


图2 作物根部甲基汞浓度变化

Figure 2 Concentrations of MeHg in root from wheat and rice after straw amendment during plant growth period

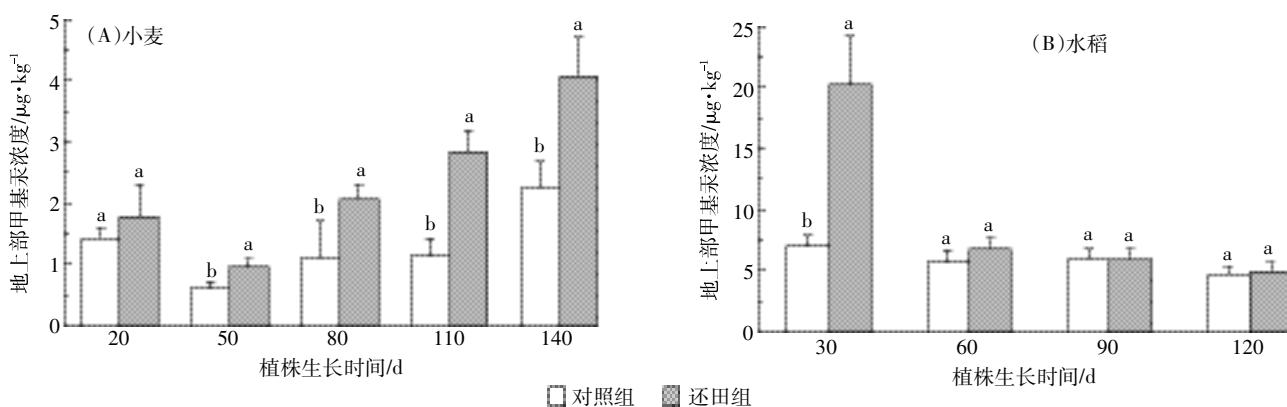


图3 作物地上部甲基汞浓度变化

Figure 3 Concentrations of MeHg in aboveground part from wheat and rice after straw amendment during plant growth period

会从茎叶向籽粒转运,使得茎叶中甲基汞含量降低。本研究也发现了类似的规律。关于小麦地上部甲基汞不断累积的原因有待进一步研究。

2.4 秸秆还田对作物籽粒甲基汞浓度的影响

研究结果表明,秸秆还田同样促进甲基汞在作物籽粒中累积(图4)。水稻秸秆还田能显著增加小麦籽粒中甲基汞的含量(图4A):秸秆还田组小麦籽粒甲基汞含量($4.09 \mu\text{g} \cdot \text{kg}^{-1}$)是对照组籽粒含量($1.41 \mu\text{g} \cdot \text{kg}^{-1}$)的2.9倍,且远低于国家食品安全限量标准 $20 \mu\text{g} \cdot \text{kg}^{-1}$ 。小麦秸秆还田组水稻籽粒甲基汞含量($16.26 \mu\text{g} \cdot \text{kg}^{-1}$)与对照组($14.79 \mu\text{g} \cdot \text{kg}^{-1}$)比较,只有略微增加,并无显著差异(图4B),但水稻籽粒中甲基汞含量接近国家食品安全限量标准。

采用籽粒甲基汞含量/土壤中甲基汞含量计算富集系数(图4C),小麦籽粒对甲基汞的累积能力远低于水稻籽粒对甲基汞的累积能力,如对照组中水稻对甲基汞的富集系数(5.46)是小麦对甲基汞的富集系数(1.04)的5.22倍。水稻籽粒甲基汞富集系数(5.78)是小麦籽粒甲基汞富集系数(0.87)的6.7倍,表明秸秆还田促进甲基汞在水稻籽粒的富集。这可能是由于秸秆还田导致土壤中甲基汞含量增加;在水稻灌浆期间,甲基汞从茎叶向稻米转运^[32],而小麦中甲基汞主要累积在茎叶中(图3A)。显然,汞矿区居民摄食稻米比摄食小麦的甲基汞暴露风险更高。

3 结论

(1)秸秆还田显著提高土壤甲基汞含量,同时增加了作物各组织甲基汞含量,这将导致汞污染地区粮食作物食品安全风险升高,因而在汞污染地区,应慎重使用秸秆还田这一传统农艺措施。

(2)不同作物秸秆还田对稻麦轮作体系土壤甲基汞及作物甲基汞的影响存在显著差别,这可能是由于变化的稻麦轮作体系与不同作物秸秆还田共同作用造成的,故秸秆还田对土壤及作物甲基汞累积的影响有待进一步研究。

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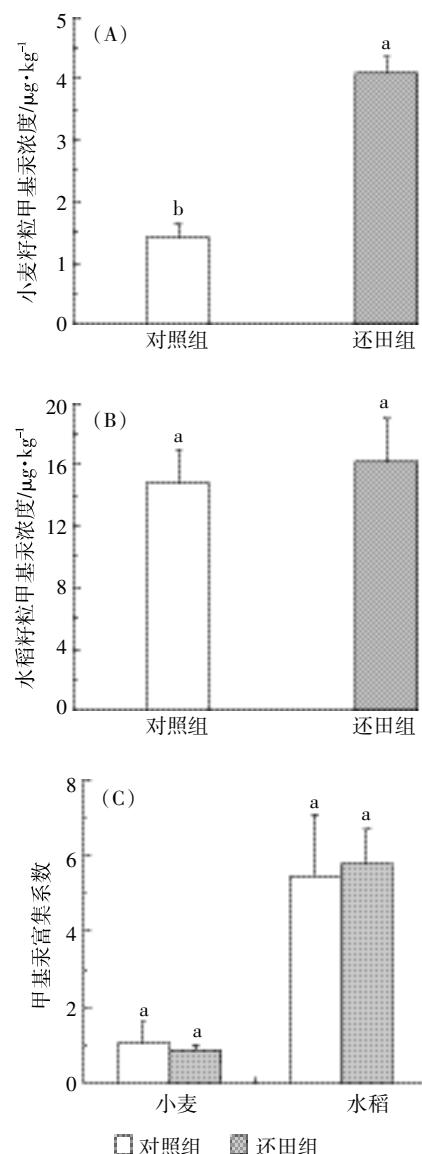


图4 作物籽粒甲基汞浓度及富集系数

Figure 4 Concentrations of MeHg in grains from wheat and rice and bioaccumulation factors of MeHg from soil to grains

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