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少化 3BA-13 枫起 MCd(ll / lh / lb / lb / lb / lb / lb / lb /	(3/4) (382) (389) (399) (405) (412)

施用污泥堆肥品对土壤和植物总汞及甲基汞的影响

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摘要:采用田间试验,分别施加两种不同的污泥堆肥品(含生物质炭和不含生物质炭),研究污泥堆肥土地利用过程土壤及植物总汞和甲基汞变化情况、迁移转化特征和植物富集能力.结果表明,施加污泥堆肥品会引起土壤总汞和甲基汞含量升高,但总汞含量低于国家土壤环境质量二级标准.生物质炭堆肥品可能促进土壤汞的甲基化,但不同处理土壤甲基汞/总汞(MeHg/THg)比值较低.植物成熟后不同处理植物总汞含量明显低于幼苗期,而甲基汞含量均高于幼苗期.施加两种堆肥品对植物富集总汞没有明显差异,但对甲基汞富集影响显著.施加生物质炭堆肥品的土壤甲基汞含量明显高于施加无生物质炭堆肥品的土壤,而植物甲基汞含量则相反.生物质炭的加入可能有利于土壤甲基汞形成,但不利于甲基汞的迁移,抑制植物对土壤甲基汞的富集.植物对甲基汞的富集能力较强(富集系数为1.24~14.63),需关注长期施肥带来的土壤环境汞生态风险.

关键词:污泥堆肥品;土地利用;植物;总汞;甲基汞;富集;生物质炭

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Effect of Sewage Sludge Compost Products Application on Total Mercury and Methylmercury in Soil and Plants

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Abstract: Two different sludge composting products (with and without biochar) were applied in field to study the variations of total mercury (THg) and methylmercury (MeHg) in soil and plants, as well as their migration in the soil-plant system and accumulation ability in plants during the sludge compost land use process. The results indicated that the concentrations THg and MeHg in soil increased after applying sewage sludge compost products, while the THg level was still lower than the secondary standard of national soil environmental quality. Biochar was speculated to probably promote the soil mercury methylation with lower MeHg/THg ratios in different soil treatments. THg concentrations in mature plants were significantly lower than those in seedling stage, but MeHg levels were higher than those in seedling stage. An obvious influence of composting on MeHg enrichment in plants was observed, and this similar effect was not found for THg enrichment. MeHg concentration in the soils applied with biochar compost was significantly higher than that without applying biochar compost soil, while MeHg in plant presented a contrary trend with higher level observed in no-biochar compost soil, suggesting that the addition of biochar could be in favour of soil MeHg formation and inhibit the MeHg accumulation in plants by influencing its migration. Since a strong MeHg accumulation ability with BCF of 1. 24-14. 63 was present in plant, the mercury ecological risk in soil environment caused by long-term fertilizing should be noticed.

Key words: sludge compost product; land use; plant; total mercury; methylmercury; accumulation; biochar

据统计,2015 年我国累计处理城镇生活污水410.3 亿 m³,而相应的污泥产量突破3 000万 t,如此巨大的污泥量已成为亟待解决、无法回避的重大城市环境问题,若处置不当则会对环境和人体健康造成严重危害。随着我国新环保法的颁布和"水十条"的实施,污泥堆肥后进行土地利用将是我国污泥处理处置与资源化的主要方式之一^[1],其富含氮、磷、钾等植物营养元素和大量的有机质等使其具有较高的利用价值。但污泥堆肥施人土壤后也可能产生二次污染,特别是增加土壤重金属含量^[2,3]。目前关于污泥土地利用重金属污染问题进行了较多的研究,郭广慧等^[4]对国内外文献(2006~2013 年)报道的污泥重金属含量进行了分析,2013 年我国城市污泥

Hg 的超标率大于 2006 年. 铁梅等^[5]通过将污泥与土壤按照一定质量比配成混合土壤进行盆栽试验,发现污泥的添加促进了黑麦草对 Cu、Cd、Zn 的吸收. 李淑芹等^[6]研究污泥堆肥对土壤和大豆不同器官的重金属积累的影响,发现随着污泥堆肥使用量的增加,土壤和大豆不同器官中 Cu、Zn、Cd 含量逐渐增加,但均未超过国家相关标准. Carbonell 等^[7]通过室内堆肥和土地利用试验发现,施加堆肥品的

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土壤 Cu、Pb、Zn含量轻微增加,而施加化肥一方面增加了土壤 Cd 和 Ni 含量,同时发现玉米果实和植物根系分别是重金属含量最低和最高的部分. 虽然关于污泥堆肥土地利用的土壤-植物体系重金属方面做了较多研究,但针对污泥堆肥土地利用过程汞及甲基汞方面的研究还很缺乏. 甲基汞作为亲脂性的有机汞,具有极高的毒性和生物可利用性,通过作物富集后进入人体会引起较大危害. 因此,本研究采用两种污泥堆肥出,分别施用到土壤中并种植作物,分析污泥堆肥土地利用过程总汞及甲基汞的迁移转化和富集特征,以期为深入开展城市污泥土地利用的重金属(特别是汞)环境风险提供基础数据.

1 材料与方法

1.1 供试材料与试验设计

试验于2015年7~8月在重庆西南大学试验大棚内进行,供试植物为耐热且早熟的不结球白菜(Brassica campestris L. ssp. chinensis Makino),试验周期35 d左右,按照传统管理模式:施肥、间苗、翻耕、灌溉等管理菜地.播种前将磨细的堆肥产品均

匀撒在各土壤样方表面后混合均匀,并浇水,后期不追肥. 种植密度为40穴·m⁻²,每穴3株,分别于发芽期第1d和第3d间苗,每天上午08:00前浇水灌溉,作物成熟后收割. 室内配备辅助照明系统,通风换气系统,透光率为75%~85%以满足植物光合作用.

所用堆肥品为自制污泥堆肥产品,按照质量比分别将污泥: 秸秆: 生物质炭 = 20: 5: 1 (A 类堆肥品)、污泥: 秸秆 = 4: 1 (B 类堆肥品)混合后放入自制堆肥箱(有效尺寸为 0. 90 m×0. 45 m×0. 71 m),采用强制通风 + 人工翻堆的方式进行好氧堆肥,堆肥产品仅限于本试验使用. 根据相关资料^[8,9],结合本次试验土壤肥力,确定土地利用试验共设 7 个处理(以施氮量计):即 CK(不施肥)、A₁、A₂、A₃(施氮量分别为 100、200、300 kg·hm⁻²)、B₁、B₂、B₃(施氮量分别为 100、200、300 kg·hm⁻²). 每块试验田为1 m²,试验田之间用 PVC 板隔开深入土壤表层 30 cm,隔绝地表径流,A、B 处理组间隔约 1 m,避免不同处理组之间相互干扰. 供试土壤和堆肥基本理化性质见表 1.

表 1 供试土壤和堆肥基本理化性质

Table 1 Basic physical and chemical properties of tested soil and sludge compost

项目	THg/mg·kg ⁻¹	MeHg/μg⋅kg ⁻¹	рН	有机质/g·kg ⁻¹	电导率/S⋅m ⁻¹	TC/g·kg ⁻¹	TN/g·kg ⁻¹
供试土壤	0.051	0. 16	7. 93	18. 10	1. 11	64. 40	1. 39
A 组堆肥	2. 203	2. 31	7. 58	550. 70	1. 83	414. 98	24. 34
B组堆肥	2. 310	1.83	7. 33	431. 03	2. 15	338. 73	24. 92

1.2 样品采集与保存

土壤样品于试验前(基本性质见表 1)、幼苗初期(2015 年 7 月 15 日)和植物收获时(2015 年 8 月 13 日)各采集一次,施肥前采集方式采用对角线采样法,幼苗期和收获期土壤样品的采集选取作物种植附近的表层土壤(0~10 cm). 土壤每次采集样品约300 g 左右,各处理组每次采集3个样品;采集的样品放入聚氯乙烯样品袋中,放入冻干机中冻干,研磨并过100 目尼龙筛后测定. 植物样的采集分别在植株幼苗期和成熟期采集各处理组所有植物样品的地上可食用部分,用去离子水冲洗干净,放入100℃烘箱烘干至恒重,称量干重,研磨并过100 目尼龙筛后装袋,于4℃条件下冷冻保存.

1.3 样品分析方法与质量控制

土壤、植物样品总汞通过 DMA-80 测汞仪进行测定,土壤和植物甲基汞采用溶剂萃取-乙基化衍生-气相色谱与冷原子荧光检测器联用方法(GC-CVAFS)测定[10,11]. pH 和电导率通过 pH 仪和电导

率仪进行测定,温度在每次采样的时候通过温度计 实时测定,有机质通过重铬酸钾容量法-加热法测 定;其他物理化学指标参照文献[12]进行测定.

试验所使用的玻璃器皿在使用前均用硝酸(25%,体积比)浸泡24h以上,超纯水清洗后经马弗炉500℃灼烧30min,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.总汞测定加标回收率为93%~105%,甲基汞测定加标回收率为89%~103%.数据处理与图形制作分别使用Excel 2010、Spss 18.0和 Origin 8.0.

2 结果与讨论

2.1 土壤总汞与甲基汞含量变化

2.1.1 土壤总汞

幼苗期土壤总汞含量变化情况见表 2. A_1 、 A_2 、 A_3 处理土壤总汞含量分别为(114.29 ± 6.61)、(135.43 ± 10.44)、(125.87 ± 1.49) μ g·kg⁻¹; B_1 、

 B_2 、 B_3 处理土壤总汞含量分别为(167.86 ± 19.27)、(174.43 ± 15.34)、(185.83 ± 12.73) $\mu g \cdot k g^{-1}$. 对比 CK,添加堆肥产品使土壤总汞含量显著升高(P < 0.05, n = 12), A_1 、 A_2 、 A_3 处理分别增加了 124.32%、165.81%、147.05%, B_1 、 B_2 、 B_3 处理分别增加了 229.46%、242.36%、264.73%.随着施肥量的增加,B 处理组土壤总汞含量逐渐升高,而 A 处理组土壤总汞含量表现为 $A_2 > A_3 > A_1$.这可能是由于 A 处理组堆肥品富含生物质炭,对施肥后汞元素在土壤中的迁移和分布产生一定影响。同时,幼苗期和收获期的土样为作物种植旁边采集,因而土壤总汞可能由于种植环境及迁移转化等条件影响产生一定差异.

到8月植物收获期,不同处理土壤总汞含量呈降低趋势.其中,A₁、A₂、A₃处理土壤总汞比7月刚试验时分别减少了6.87%、5.50%、11.97%,B₁、B₂、B₃处理分别降低了20.19%、8.73%、11.63%,这可能是由于土壤中汞的转化、释放和植物对土壤汞元素的吸收所致.分析发现,施加污泥堆肥产品虽然能增加土壤总汞含量,但都低于国家土壤环境质量二级标准限值(GB 15618-1995),没有引起土壤汞污染,但若长期施用可能会造成土壤汞累积,存在潜在的汞生态风险.

2.1.2 土壤甲基汞变化特征

土壤甲基汞含量变化情况如图 1. 结果显示,幼苗期(7月15日) A_1 、 A_2 、 A_3 处理土壤甲基汞含量分别为 0. 28、0. 40、0. 38 $\,\mathrm{ng}\cdot\mathrm{g}^{-1}$, B_1 、 B_2 、 B_3 处理则分别为 0. 23、0. 22、0. 38 $\,\mathrm{ng}\cdot\mathrm{g}^{-1}$, 其含量与一般旱地土壤甲基汞含量相近,甚至低于稻田土壤 $^{[13,14]}$, A组土壤甲基汞含量总体上略高于 B 组.对比 CK,施加污泥堆肥品使土壤甲基汞含量明显升高, A_1 、 A_2 、 A_3 和 B_1 、 B_2 、 B_3 土壤甲基汞升高量分

别为 75. 45%、151. 33%、135. 29% 和 45. 47%、38. 66%、139. 14%,A 处理组土壤甲基汞增加量总体高于 B 处理组. 随着施肥量的增加,两组处理土壤甲基汞含量没有明显变化趋势. 试验结束时(8月13日), A_1 、 A_2 、 A_3 和 B_1 、 B_2 、 B_3 土壤甲基汞含量分别为 0. 34、0. 49、0. 39 $\operatorname{ng} \cdot \operatorname{g}^{-1}$ 和 0. 21、0. 10、0. 19 $\operatorname{ng} \cdot \operatorname{g}^{-1}$. 图 1(b)可以看出,A 处理组土壤甲基汞含量明显高于 B 处理组(P < 0.05, n = 12).

表 2 不同处理土地利用土壤总汞含量变化1)/µg·kg-1

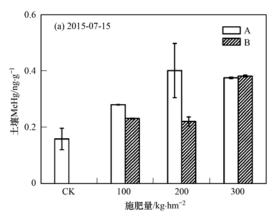
land was pressed in a / was beg-1

Table 2 Total mercury in the soil of different

	land use processing/ μg·κg	5 -
处理组	2015-07-15	2015-08-13
CK	50.95 ± 1.42a	56.72 ± 0.57a
\mathbf{A}_1	114.29 ± 6.61 b	$106.44 \pm 8.97 \mathrm{b}$
\mathbf{A}_2	$135.43 \pm 10.44 d$	$127.98 \pm 7.77 d$
A_3	$125.87 \pm 1.49c$	$110.80 \pm 8.00c$
B_1	$167.86 \pm 19.27e$	$133.97 \pm 0.85e$
B_2	174.43 ± 15.34 f	$159.21 \pm 5.41f$
B_3	185.83 ± 12.73 g	$164.22 \pm 6.32g$

1)同列数值的不用字母表示 Duncan's 多重比较差异显著 (P < 0.05, n = 12)

与幼苗期相比,试验结束时 A 组 3 种处理的甲基汞含量均有不同程度增加,而 B 组 3 种处理土壤甲基汞含量则有所降低. 分析原因,可能是由于 A 组处理加入了含生物质炭的堆肥品,提高了土壤有机质含量. Porvari 等[15]发现有机质作为甲基化微生物的营养物质,能够为其提供充足碳源,提高微生物活性,促进汞甲基化. Graber等[16]也认为施加生物质炭能够增加微生物群落,促进土壤团聚体的形成,改善土壤物理性质,保持水气协调,为微生物的活动创造了良好的条件. 因此施加生物质炭堆肥品的 A 处理组土壤甲基汞含量高于 B 处理组. Weber^[17]研究发现 DOM 能促进汞的甲基化,而



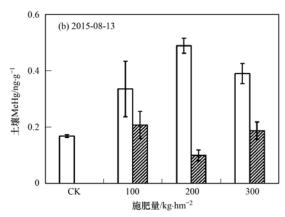


图 1 土壤甲基汞含量变化情况

Fig. 1 Content changes of soil methylmercury

Barkay 等^[18]却认为 DOM 的络合作用能够提高汞溶解和抑制汞形成硫化物沉淀,限制甲基化细菌对Hg²⁺的可利用程度和甲基汞的生物累积效率.本试验也发现,生物质炭堆肥品加入后,增加了土壤有机质含量,土壤甲基汞含量增加,但施肥量超过一定范围后,土壤甲基汞含量出现降低,表明有机质对土壤汞的甲基化可能具有双重作用,但其影响机制需要进一步研究分析.

2.1.3 土壤 MeHg/THg 比值

分析发现,不同处理土壤 MeHg/THg 比值较 低. 刚施入污泥堆肥品时, A,、A,、A,处理和 B,、 B₂、B₃处理土壤 MeHg/THg 比值分别为 0.25%、 0.30%、0.30%和0.14%、0.13%、0.21%, CK处 理 MeHg/THg 比值为 0.31%,和湖泊和湿地沉积物 MeHg/THg 比值相近[19]. 本试验结束时,土壤 MeHg/THg 比值分别为 0.32%、0.38%、0.35% 和 0.16%、0.06%、0.11%, CK 处理 MeHg/THg 比值 为 0.30%. 总体来说, A 处理组 MeHg/THg 比值高 于 B 处理组,这可能与土壤中甲基汞含量较高以及 A 组处理含有生物质炭有关. 有研究表明土壤汞主 要以性质稳定的残渣态形式存在[20~22],活性汞含量 极少. 彭倩等[23] 发现腐殖酸的加入导致土壤中有 机结合态汞含量显著上升,说明无机汞与腐殖酸存 在络合作用,Cunha-Santino 等[24] 也发现有机质对 土壤重金属的矿化作用,从而降低对土壤微生物的 生物有效性,因此本试验中土壤汞甲基化率均处于 较低水平.

2.2 植物总汞和甲基汞含量变化

2.2.1 植物总汞

表 3 结果显示,植物幼苗期 A_1 、 A_2 、 A_3 处理植物总汞含量分别为(314.99 ± 63.26)、(377.77 ± 111.13)、(321.45 ± 19.03) $\mu g \cdot k g^{-1}$; B_1 、 B_2 、 B_3 处理植物总汞含量分别为(397.87 ± 46.85)、(429.22 ± 80.30)、(377.15 ± 173.21) $\mu g \cdot k g^{-1}$, A、B 各处理组内植物总汞含量无显著差异。而到 8 月收获期,植物总汞含量为出现不同程度降低,其中 A_1 、 A_2 、 A_3 处理分别下降了 60.14%、44.93%、38.76%; B_1 、 B_2 、 B_3 处理分别下降了 52.91%、58.10%、55.25%。这主要是由于污泥堆肥品富含有机质和植物所需多种微量元素,在幼苗期植物生长需要大量养分,根系吸收能力较强,从施污土壤中吸收养分的同时,富集了大量的汞。同时,植物中的汞也可以通过气孔吸收 Hg^0 、叶片吸附大气 Hg^0 、 Hg^{2+} 以及通过蒸腾作用吸收土壤可溶性汞[25.26].

而收获期 90% 以上的植物已成熟,需要的养分较少,植物体内生长代谢、化学转化等过程逐渐减少,基本不再从土壤中吸收营养,植物体内吸收的生物可利用汞含量也基本稳定,因而收获期植物汞含量低于幼苗期. A_1 、 A_2 、 A_3 处理和 B_1 、 B_2 、 B_3 处理的植物总汞含量无明显差异(P>0.05,n=12),说明生物质炭的添加对植物总汞的吸收未产生显著影响.

表3 不同处理植物总汞含量/µg·kg-1

Table 3 $\,$ Total mercury in the plants of different land

	use processing/µg∙kg - 1	l.
处理组	2015-07-15	2015-08-13
CK	167.42 ± 72.44b	121.32 ± 27.15d
\mathbf{A}_1	$314.99 \pm 63.26a$	$125.55 \pm 11.81\mathrm{bc}$
A_2	377.77 ± 111.13a	$208.03 \pm 10.70a$
A_3	$321.45 \pm 19.03a$	$196.84 \pm 34.91a$
B_1	$397.87 \pm 46.85a$	$187.35 \pm 44.66a$
B_2	$429.221 \pm 80.30a$	$179.84 \pm 37.12ab$
B_3	377.15 ± 173.21a	168.78 ± 25.37 abc

2.2.2 植物甲基汞

植物甲基汞含量变化见图 2. 幼苗期 A 处理组 植物甲基汞含量范围为 0.21~0.54 ng·g⁻¹, B 处理 组植物甲基汞含量范围为 0.28~1.13 ng·g-1,除 B3 处理外,其余处理均高于 A 处理组. 收获期不同处 理植物体内甲基汞含量比幼苗期均有不同程度增 加,A组增加幅度较小,而B组B,、B,处理分别增 加了1.4和3.7倍. A处理组植物甲基汞含量显著 低于 B 处理(P < 0.05, n = 12),与土壤甲基汞含量 正好相反(A处理组土壤甲基汞含量明显高于 B处 理组),说明生物质炭对植物甲基汞的吸收或者土 壤甲基汞自身迁移能力造成一定抑制. 虽然试验中 生物质炭的加入增加了土壤有机质含量(土壤有机 质均值从 1.81% 增加到 6.11%),但有机质的增加 特别是腐殖酸与汞具有矿物质结合、络合反应等吸 附作用,从而使其在土壤中被固定下来,导致其甲基 化后难以从土壤迁移到植物体内.

2.3 植物汞富集水平

植物中重金属的富集系数 (bioconcentration factors, BCFs) 可以用植物中的重金属含量与土壤重金属含量的比值来表示^[27]. 不同处理植物汞的富集系数见图 3. 从中可知, 植物对甲基汞的富集系数为 1. 24 ~ 14. 63, 显著高于(P < 0. 05, n = 12) 总汞的富集系数 (1. 03 ~ 1. 78), 其结果与梁丽等^[28]的研究结果相近. 本试验在大棚里面进行, 受大气干湿沉降的影响很小, 植物富集的汞一方面来源于土壤, 另

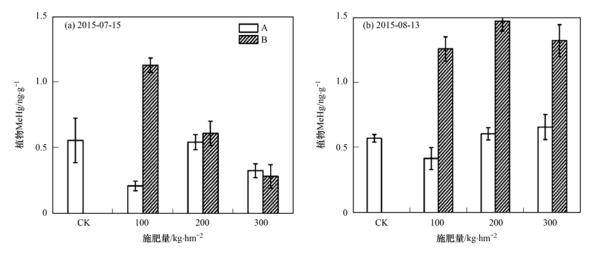


图 2 植物甲基汞含量变化

Fig. 2 Content changes of plant methylmercury

外也可能通过植物叶片吸收大气中的汞以及通过蒸腾作用吸收土壤释放的汞^[29,30].有研究发现,水稻地上组织中的汞主要来自对大气汞的吸收,茎部和叶部少部分汞来自根系对土壤汞的吸收^[26],表明大气汞对植物汞含量也有一定影响.但由于仪器设备的限制,本次试验未对大气汞浓度进行监测.研究还发现,A、B 两处理组植物对总汞的吸收没有明显差异(P>0.05,n=12),但 A 组甲基汞富集系数显著低于 B 组(P<0.05,n=12),表明生物质炭的加入可能降低植物对土壤甲基汞的富集,这与前面对植物体内甲基汞含量分析结果一致,但其影响机制还需进一步研究.

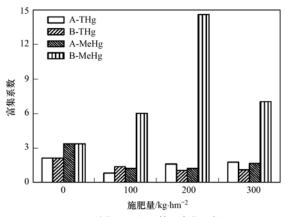


图 3 植物总汞和甲基汞富集系数

Fig. 3 BCFs of THg and MeHg in the plants

2.4 土壤-植物系统汞迁移转化影响因素

汞在土壤中的积累、迁移和转化受土壤体系中的生物、物理过程和氧化还原、沉淀溶解、吸附解吸、络合螯合和酸碱反应等复杂化学过程的影响^[31],因此对土壤理化性质和土壤、植物总汞及甲

基汞进行相关性分析. 结果显示,A 处理组土壤甲 基汞与有机质呈显著正相关关系(R = 0.955*,P =0.045, n = 12). 本试验中 A 处理组污泥堆肥品富含 生物质炭导致土壤有机质含量升高,有研究称有机 质可以促进土壤汞甲基化[32],因此在刚施入污泥堆 肥品和试验结束时,A处理土壤甲基汞含量明显高 于 B 处理,说明生物质炭的添加可能有利于土壤汞 甲基化. 此外,生物质炭的加入可能改变了土壤内 部环境,其多孔性和表面特性可以为土壤微生物提 供附着位点和较大的空间,并且其保水保肥的能力 对微生物的数量和活性起促进作用[33,34],从而有利 于甲基化微生物活动,进而影响到土壤甲基汞的含 量. 此外,B处理组植物甲基汞与TC 呈显著正相关 关系(R = 0.961 * , P = 0.039, n = 12), TN、pH 等其 他因素与土壤、植物总汞及甲基汞之间相关性均不 明显.

3 结论

- (1)施加污泥堆肥产品能增加土壤总汞和甲基汞含量,但总汞低于国家土壤环境质量二级标准. 生物质炭堆肥品的加入可能促进土壤中汞的甲基化,增加土壤甲基汞含量,不同处理土壤 MeHg/THg 比值均处于较低水平.
- (2)植物幼苗期富集了大量的汞,收获期植物总汞含量低于幼苗期,而甲基汞含量均高于幼苗期, 且施加生物质炭堆肥品对土壤甲基汞的迁移和植物对甲基汞的吸收具有一定抑制作用.
- (3)植物对甲基汞富集能力显著高于总汞,污 泥堆肥品的长期施用可能会造成潜在的汞生态风

险. 生物质炭对植物总汞的富集未产生显著影响, 但降低了植物对土壤甲基汞的富集.

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