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生物滴滤塔净化甲基叔丁基醚废气的研究

褚其英¹,姚露露¹,吕雄标²,叶杰旭¹,叶虹霓¹,潘梁柱¹,陈建孟¹,陈东之¹*

(1. 浙江工业大学环境学院, 杭州 310032; 2. 浙江菲尔特环保工程有限公司, 杭州 310014)

摘要:应用生物滴滤塔处理甲基叔丁基醚废气,研究其挂膜启动及稳定运行阶段的降解性能,并考察了稳定期该系统的生物群落结构.结果表明,生物滴滤塔在停留时间为60 s,进气质量浓度为100 mg·m⁻³的条件下,运行23 d 后完成挂膜,填料上的生物量明显增加,去除率可维持在70%以上.反应器稳定运行时,去除负荷可达13.47 g·(m³·h)⁻¹,矿化率可达68%;用 Haldane 模型拟合生物滴滤塔中去除负荷的变化趋势,获得理论 EC_{max} 为21.03 g·(m³·h)⁻¹, K_s 为0.16 g·m⁻³, K_l 为0.99 g·m⁻³.运用高通量测序技术分析生物膜中的微生物群落结构,发现其中优势菌属为 Methylibium sp.和 Blastocatella sp.,分别占11.33%和9.95%.

关键词:生物滴滤塔;甲基叔丁基醚;去除负荷;动力学

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Treatment of the Waste Gas Containing Methyl *tert*-Butyl Ether via a Biotrickling Filter

CHU Qi-ying 1 , YAO Lu-lu 1 , LÜ Xiong-biao 2 , YE Jie-xu 1 , YE Hong-ni 1 , PAN Liang-zhu 1 , CHEN Jian-meng 1 , CHEN Dong-zhi 1 *

(1. College of Environment, Zhejiang University of Technology, Hangzhou 310032, China; 2. Feierte Environmental Engineering Company Limited of Zhejiang Province, Hangzhou 310014, China)

Abstract: The performance and microbial communities of methyl *tert*-butyl ether (MTBE) treatment using a biotrickling filter (BTF) that was inoculated with activated sewage sludge were investigated. The BTF successfully started up within 23 days when the inlet concentration of MTBE was 100 mg·m⁻³ and empty bed retention time was 60 s, with 70% removal efficiency (RE). Under steady-state conditions, an elimination capacity (EC) and a mineralization ratio of 13.47 g·(m³·h)⁻¹ and 68% were achieved, respectively. The EC_{max} was 21.03 g·(m³·h)⁻¹ according to the Haldane model, and a K_s of 0.16 g·m⁻³ and K_l of 0.99 g·m⁻³ were obtained. High-throughput sequencing was used to identify the community structure of the mixed microbial consortium in the BTF. The results indicated that *Methylibium* sp. (11.33%) and *Blastocatella* sp. (9.95%) were the dominant bacteria.

Key words: biotrickling filter; methyl tert-butyl ether (MTBE); elimination capacity; kinetic

甲基叔丁基醚(MTBE)是一种无色、透明、高辛烷值的液体,作为无铅汽油的添加剂,已经在世界各地广泛使用^[1,2]. MTBE 具有水溶性高、又能与其他有机污染物共溶的特性,使其他污染物在水中的溶解度提高 1 个数量级,造成更严重的化学污染. 大量毒理学研究则表明^[3,4], MTBE 是一种动物致癌物质,同时也会影响人体的中枢神经,且是一种人体可疑致癌物质. 美国国家环保署(USEPA)已将其列在环境优先污染物的名单当中^[5].

相关研究发现^[6,7],化学方法处理 MTBE 具有较好的效果. 但因为 MTBE 正辛醇-水分配系数小,与土壤的结合能力弱,限制了吹脱、高级氧化、活性炭吸附等物理化学处理工艺的应用^[2]. 相比上述物理化学方法,生物法可利用微生物的代谢活动将有机污染物转化为 CO₂、H₂O 以及生物质能,因此适用于大气量、低浓度的有机废气的净化,是一种

经济、安全、高效的处理技术^[8]. 然而由于 MTBE 的碳链较短, 使得微生物对碳源的利用受到抑制, 生物量积累期长, 从而造成 MTBE 的生物降解速率比一般有机物的降解要慢^[9]. 不同研究报道的 MTBE 生物降解周期差异很大, 有些能在几天内完成对 MTBE 的降解^[2,10], 但有些需要十几天甚至几十天^[11,12], 并且根据环境条件的不同, MTBE 的最终降解率差别很大^[13,14]. 因此, 探索 MTBE 废气的生物净化工艺具有重大的理论和实践意义.

目前主要的生物处理 VOCs 的工艺有:生物过滤法^[12],生物洗涤法^[15],生物滴滤法^[11]等.其中的生物滴滤法具有处理能力大、适用范围广、运

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作者简介: 褚其英(1992~), 女, 硕士, 主要研究方向为环境生物

技术, E-mail:cqy@zjut.edu.cn

* 通信作者,E-mail:cdz@zjut.edu.cn

行工况易于调节、填料不易堵塞等优点,有利于生物净化法的工程推广,因此备受研究者的关注^[16-18].本研究利用生物滴滤塔净化 MTBE 废气,考察生物滴滤塔对 MTBE 的去除能力,并用宏观动力学模拟其去除负荷随进口浓度的变化趋势,为工程化生物滴滤塔处理 MTBE 废气奠定理论基础;同时考察反应器运行阶段生物量的变化情况,并利用高通量测序解析复杂的生物群落结构,以期为进一步利用生物滴滤塔处理 MTBE 废气奠定基础.

1 材料与方法

1.1 实验装置及流程

本实验生物滴滤塔主要由设有废气进口的塔底、设有生物填料层、取样口和水浴保温夹套的塔身、安装有尾气出口、气体采样口和营养液喷淋系统的塔顶、空气泵、吹脱瓶、混合瓶组成;营养液喷淋系统由安装在塔顶的喷洒器,设在生物滴滤塔外部的循环营养液储存瓶、营养液输入管、pH控制仪、蠕动泵、碱液瓶连接组成;pH控制系统由pH控制仪分别与循环营养液储存瓶和碱液瓶连接。实验采用逆流式操作,空气经空气泵、质量流量计后进入装有液态MTBE的吹脱瓶中,将MTBE废气吹出,废气经胶管到达气体混合瓶与空气充分混合,混合气体经流量计调节流量后,模拟废气从塔底进

入生物滴滤塔,被生物滴滤塔中附着在填料上的生物膜净化.实验装置如图1所示.

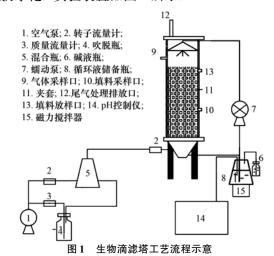


Fig. 1 Schematic diagram of the biotrickling filter

生物滴滤塔由总高 620 mm、内径 130 mm 的有机玻璃制成,填料层高度为 300 mm,沿塔高方向设置 2 个气体采样口(1 个进气口,1 个出气口),1 个填料取样口,1 个填料放样口.实验采用气液逆流操作,气体由塔底进入,由下而上,营养液经蠕动泵从循环液储备瓶提升至塔顶向下喷淋,储液瓶容积为 1.5 L,喷淋速率为 180 mL·min⁻¹. 挂膜完成后,装置设有 pH 自动控制系统,pH 维持在 6.9~7.2,每1 d 更换 0.5 L 营养液,稳定运行阶段每日更换的营养液中 MTBE 的含量具体如表 1 所示.

表 1 不同进气质量浓度下每日更换的营养液中 MTBE 的含量

	1.09	,						
Tabla 1	Content of	MTDE for	the overla	aalusian	zwiela ela	. different	inlat	concentrations
rabie i	Content of	MIIDE 101	the evere	Solution	WILLI LITE	: amerem	шиец	concentrations

序号	MTBE 进气质量浓度 /mg·m ⁻³	流经生物滴滤塔 MTBE 总量 /mg·d ⁻¹	更换的营养液中 MTBE 含量 /mg·d -1
1	103	593. 3	1. 1
2	179	1 031. 0	2. 5
3	511	2 943. 4	5. 2

1.2 填料与营养液

聚氨酯小球具有比表面积大、空隙率较大、压降小、抗老化力强等特征,因此采用聚氨酯小球作为生物滴滤塔中的填料,具体参数见文献[8].

采用连续喷淋的方式向生物滴滤塔中提供营养液,其组分如下: $CaCl_2$ 0. 03 $g \cdot L^{-1}$, $MgSO_4 \cdot H_2O$ 0. 46 $g \cdot L^{-1}$, $(NH_4)_2SO_4$ 1. 23 $g \cdot L^{-1}$, KH_2PO_4 0. 7 $g \cdot L^{-1}$, K_2HPO_4 0. 85 $g \cdot L^{-1}$, $FeSO_4 \cdot 7H_2O$ 0. 001 $g \cdot L^{-1}$, 微量元素母液 1 $mL \cdot L^{-1}$; 溶剂为水,pH 7. 0 ~ 7. 5;

微量元素母液:FeSO₄·7H₂O 1.0 g·L⁻¹, CuSO₄·5H₂O 0.02 g·L⁻¹, H₃BO₃ 0.014 g·L⁻¹, MnSO₄·

$$\begin{split} 4 H_2 O & 0. \ 10 \quad g \cdot L^{-1} \ , \quad Zn SO_4 \ \cdot \ 7 H_2 O \ 0. \ 10 \quad g \cdot L^{-1} \ , \\ N a_2 MoO_4 \ \cdot \ 2 H_2 O \ 0. \ 02 \quad g \cdot L^{-1} \ , \quad CoCl_2 \ \cdot \ 6 H_2 O \ 0. \ 02 \\ g \cdot L^{-1} . \end{split}$$

1.3 分析方法

1.3.1 气相色谱法(GC)

MTBE 检测分析方法:采用 Agilent 6890 气相色 谱仪(Agilent, 美国), 色谱柱为 HP-Innowax 硅胶毛细管柱(30 m × 0.32 mm × 0.5 μm). 气相色谱条件:进样口、检测器温度(FID)和柱温分别为250℃、80℃和300℃,柱流量为1 mL·min⁻¹,分流比为5:1,进样体积为400 μL.

CO, 定量检测: 采用 Agilent 6890 气相色谱仪

(Agilent, 美国), 色谱柱为 HP-Plot-Q 毛细管柱(30 m×0.32 mm×20 μ m). 气相色谱条件:进样口、检测器温度(TCD)和柱温分别为90、100 和40℃, 柱流量为5 mL·min⁻¹, 进样体积为1000 μ L, 尾吹气: 氦气. **1.3.2** 生物量的测定

蛋白质含量测定:从填料采样口取 1 枚聚氨酯 小球,放入 50 mL 离心管中,加入 15 mL 无菌水漩 涡振荡 10 min;取一定的菌悬液超声破碎,离心后获得的上清液即为所需检测的蛋白质;采用考马斯亮蓝染色法测定其中蛋白质浓度.

高通量测序:由生工生物工程(上海)股份有限公司完成. 测序区域选择 V3-V4 区, 测序片段为465 bp, 测序引物为341F(CCTACGGGNGGCWGCAG)-805R(GACTACHVGG-GTATCTAATCC),并选择细菌为研究对象.

2 结果与讨论

2.1 BTF 的挂膜启动

活性污泥取自浙江某制药厂的曝气池,于实验室条件,通入一定量的 MTBE 曝气培养 15 d. 测试得 SV 值为 53%, MLSS 为 $3590~{\rm mg\cdot L^{-1}}$, SVI 为 $148~{\rm mL\cdot g^{-1}}$.

为避免不必要的实验误差,反应器在接种活性污泥进行挂膜前,预先通人质量浓度为 100 mg·m⁻³的 MTBE 模拟废气,待反应器进出口的MTBE 质量浓度基本一致,即填料吸附饱和,后按活性污泥:营养液体积比1:2的接种方式,通过喷淋循环系统进行喷淋挂膜. 挂膜期,生物滴滤塔的停留时间控制为 60 s, MTBE 进气质量浓度维持在100 mg·m⁻³.

目标污染物的去除能力是衡量生物滴滤塔启动效果的重要指标.启动阶段,生物滴滤塔对 MTBE 的去除情况如图 2 所示. 挂膜 23 d 后, MTBE 的去除率达到 70%以上,并能保持稳定的去除能力,该结果表明生物滴滤塔完成挂膜. Sercu 等[19] 用生物滴滤塔治理甲硫醚废气,分别以单一的活性污泥,活性污泥+高效菌进行接种挂膜,两者都在运行的10 d 后完成挂膜,挂膜所需时间较短,可能与反应器挂膜时,进气底物浓度较低有关. 本实验只用活性污泥挂膜处理 MTBE 废气,挂膜完成时间较长,对 MTBE 去除能力较弱,也可能与底物性质有关. 相关研究表明[20,21], MTBE 的碳链较短,影响微生物对碳源物质的利用能力,导致生物降解速率比一般有机物慢.

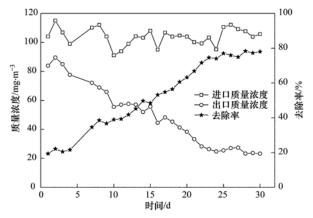


图 2 生物滴滤塔挂膜阶段 MTBE 进出口浓度和去除率

Fig. 2 Concentration and removal efficiency of MTBE during the start-up phase of the BTF

2.2 进气质量浓度对去除率和去除负荷影响

进气质量浓度影响生物滴滤塔的降解性能. 因 此,考察停留时间(EBRT)为60 s时,不同进气质 量浓度的 MTBE 对去除率及去除负荷的影响,结果 如图 3 所示. 随着 MTBE 进口浓度从 100 mg·m⁻³升 高到 560 mg·m⁻³, 去除率从 75% 下降到 30%; 随 着进口负荷的增大,去除负荷从5.11 g·(m³·h)-1 逐步升高到13.47 g·(m³·h)-1, 当进气负荷进一步 增大到 33.56 g·(m³·h)-1 时,去除负荷下降到 11.06 g·(m³·h) -1. 结果表明, 随着进气质量浓度 的增加, 生物滴滤塔对 MTBE 的去除率下降. 而去 除负荷先升高后下降的现象,则是因为在相对较高 的进气负荷条件下,单位生物量产生的单位酶结合 底物能力趋向饱和, 而进一步增加进气负荷, 可能 抑制了反应器体系内的生物活性,从而使生物滴滤 塔的去除能力下降^[8]. Eweis 等^[22]报道的生物过滤 器在 EBRT 为 60 s 时,对 MTBE 的去除负荷也仅为 8 g·(m³·h)⁻¹, 比本实验去除负荷能力差. Nikpey 等[23]报道的生物滴滤塔对 MTBE 的去除负荷能达 到 25 g·(m³·h) -1, 该文中也明确表示当仅向生物

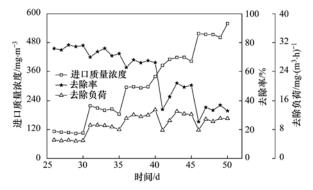


图 3 进气质量浓度对 MTBE 去除率的影响

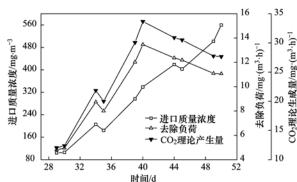
Fig. 3 Effects of inlet concentration on removal rate

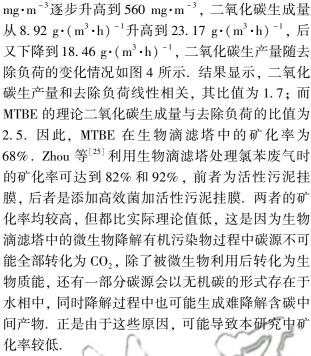
滴滤塔中接种土著微生物时,运行 13 个月都未能实现对 MTBE 的净化,可见后期接种的具有 MTBE 降解性能的微生物组起到了明显净化 MTBE 的作用.不同研究报道的 MTBE 生物降解周期以及降解能力差异很大^[2,10~14].由此可见,MTBE 降解微生物的种类以及单位体积该微生物的含量都会影响MTBE 的降解能力.

2.3 二氧化碳生成量分析

在生物降解过程中,有机污染物能够被转化为 H₂O、CO₂ 和生物质能.因此,二氧化碳生成量是 衡量生物滴滤塔的去除性能的一个重要指标.一些 研究中还经常涉及二氧化碳理论生成量这个概 念^[24,25],即有机污染物中的碳源完全转化为二氧化碳.而二氧化碳实际生成量和理论生产量的比值——矿化率,也是一个衡量或比较生物滴滤塔性能的重要参数.

本实验中, MTBE 的进气质量浓度从 100





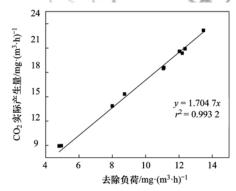


图 4 CO₂ 生成量随去除负荷的变化关系

Fig. 4 Relationship between CO2 production rate and elimination capacity

2.4 生物滴滤塔动力学分析

生物滴滤塔中去除负荷的变化趋势,可以用宏观动力学方程来描述. Michaelis-Menten 模型^[24]是一种常用的描述反应器动力学过程的方程,其表达式如下:

$$EC = \frac{EC_{\text{max}}C_{\text{ln}}}{K_{\text{S}} + C_{\text{ln}}}$$

式中, EC 为去除负荷, $g \cdot (m^3 \cdot h)^{-1}$; C_{ln} 为进、出口浓度的对数平均值, $g \cdot m^{-3}$; EC_{max} 为最大去除负荷, $g \cdot (m^3 \cdot h)^{-1}$; K_s 为饱和常数, $g \cdot m^{-3}$.

当进气质量浓度超过一定值后,去除负荷受到抑制时,常用 Haldane 模型^[26]对实验数据进行拟合,其表达式如下:

$$EC = \frac{EC_{\text{max}1} C_{\text{ln}}}{K_{\text{S1}} + C_{\text{ln}} \left(1 + \frac{C_{\text{ln}}}{K_{\text{I}}}\right)}$$

式中, EC_{max1} 为不存在底物抑制时的最大去除负荷, $g \cdot (m^3 \cdot h)^{-1}$; K_{S1} 为饱和常数, $g \cdot m^{-3}$; K_{I} 为抑制常数, $g \cdot m^{-3}$.

分别用 Michaelis-Menten 和 Haldane 模型拟合 去除负荷随 C_{ln} 从 $0.037 \sim 0.537~g\cdot m^{-3}$ 的变化趋势,结果如图 5 所示. 当 C_{ln} 大于 $0.3~g\cdot m^{-3}$ 时,去除负荷下降,同时对比图 5,显然拟合结果 r^2 为 0.91 的 Haldane 模型拟合数据的效果更好. Haldane 模型拟合获得的 EC_{max} 为 $21.03~g\cdot (m^3\cdot h)^{-1}$, K_s 为 $0.16~g\cdot m^{-3}$, K_l 为 $0.99~g\cdot m^{-3}$,该结果显示当 C_{ln} 大于 $0.99~g\cdot m^{-3}$ 时,存在底物抑制现象. 由图 3 也可发现,当进口浓度大于 $300~mg\cdot m^{-3}$ 后,MTBE 的去除率和去除负荷随着 MTBE 的浓度升高而下降. 该现象同多数的序批式纯培养实验,即高底物浓度抑制微生物的活性 $[^{2.27}]$. 而 Lin 等 $[^{28}]$ 在生物滴滤塔处理

MTBE 废气实验中,用 Michaelis-Menten 模拟获得的 EC_{max} 为 22.7 $g \cdot (m^3 \cdot h)^{-1}$, K_s 为 0.10 $g \cdot m^{-3}$. Gallastegui 等 $[^{24}]$ 在生物滴滤塔净化甲苯废气实验中,发现当 C_{ln} 大于 3 $g \cdot m^{-3}$ 时,存在底物抑制现象,因此认为 Haldane 模型比 Michaelis-Menten 模型拟合性更好。由于反应器尺寸大小、活性污泥特性及底物性质不同,底物抑制浓度各不相同 $[^{25,26,29}]$,造成底物的降解机制不同,同时也促使生物滴滤塔适用的模型各有差异.

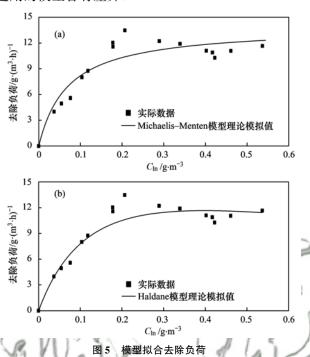


Fig. 5 EC curve fitting with different models

2.5 微生物相分析

生物滴滤塔中的生物量是评价反应器性能的一个重要指标^[8],一般研究采用蛋白质含量来表征并定量生物滴滤塔中的生物量. 在生物滴滤塔运行过程中,选取不同时间段的填料样品,采用考马斯亮蓝染色法测定其中的蛋白质含量,结果如图 6 所示. 反应器启动挂膜 5、10、20、30 d 后,测定蛋白质含量分别为 0. 19、0. 55、0. 83、0. 97 mg·g⁻¹. 蛋白质含量逐渐趋于稳定,说明生物滴滤塔完成挂膜. 而在生物滴滤塔稳定运行阶段,蛋白质含量稳定维持在 1.0 mg·g⁻¹左右,则说明生物膜的脱落和更新速度达到动态平衡,也说明该阶段微生物具有良好的降解能力.

生物滴滤塔填料上的生物膜中包括可以直接降解污染物的微生物,以及捕食细菌和吞噬有机颗粒的原生动物、后生动物.复杂的生物膜群落结构有助于生物滴滤塔系统的物质转化,并能稳定生态系

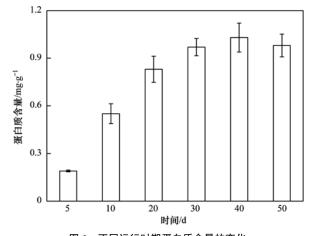


图 6 不同运行时期蛋白质含量的变化

Fig. 6 Protein quantity versus operation time

统,保证稳定废气的去除能力.为分析生物滴滤塔中生物膜中的微生物群落结构,对其进行了宏基因组微生物分类测序.取反应器完成挂膜后(30 d)的填料,进行高通量测序,门水平和属水平的优势菌组成鉴定结果如图 7 所示.

由图 7(a) 可知, 生物滴滤塔中最大的优势细菌门为变形菌门(Proteobacteria), 占总细菌序列的34%, 该结果同 Snaidr 等^[30] 对常规活性污泥中群落多样性的研究. 变形菌门中包含多种能分解有机物的代谢菌^[31,32], 这可能是其大量存在的原因. 由图 7(b) 可以看出 *Methylibium* sp. 和 *Blastocatella* sp. 所占比例较高,分别为 11.33%和 9.95%.

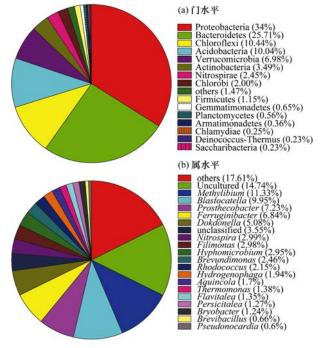


图7 菌群群落结构(门和属水平)

Fig. 7 Bacterial community structure at the phylum and genus levels

Methylibium 属细菌常具有良好的 MTBE 降解能力,如 M. petroleiphilum PM1^[31]、Methylibium sp. strain T29^[33]、Methylibium sp. R8^[34]等. Blastocatella 属细菌被发现能够水解有机物^[35]. 分别占 2.15%和1.7%的 Rhodococcus 属和 Aquincola 属细菌,也被发现能够降解 MTBE^[36,37]. 因混合菌群中同时存在多种具有 VOCs 污染物降解能力的功能菌,使得生物滴滤塔具有良好及稳定的 MTBE 降解能力.

3 结论

- (1)制药厂活性污泥经前期驯化后,接种到生物滴滤塔中用于净化低浓度的 MTBE 废气.在EBRT为60 s,进气质量浓度为100 mg·m⁻³的条件下,挂膜所需时间为23 d, MTBE的去除率在70%以上.
- (2) 生物滴滤塔稳定运行阶段,随着进气质量浓度的增加去除负荷先上升后下降,MTBE的最大去除负荷为13.47 g·(m³·h)⁻¹,同时 MTBE 的矿化率可达68%.由于高进气质量浓度造成去除负荷下降,因此存在底物抑制,生物滴滤塔去除 MTBE 废气符合 Haldane 模型.理论 EC_{max} 为 21.03 g·(m³·h)⁻¹,K_s 为 0.16 g·m⁻³,K₁ 为 0.99 g·m⁻³.
- (3) 生物滴滤塔中优势菌属主要为 Methylibium sp. 和 Blastocatella sp., 所占比例分别为 11.33% 和 9.95%.

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