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# 巫山黄土微生物四醚膜脂分布特征及其古气候意义

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**摘要:** "巫山黄土"是指分布于长江三峡地区的黄土状堆积物,是中国黄土的重要组成部分,具有重要的古气候环境意义,微生物四醚膜脂是反演古气候环境的灵敏指标,为了进一步理解巫山黄土中蕴含的古气候意义,通过测定巫山黄土中微生物醚类化合物(glycerol dialkyl glycerol tetraethers, GDGTs)并分析其分布特征,基于支链 GDGTs(bGDGTs)的甲基化指数(methylation index of branched tetraethers, MBT)和环化指数(cyclization ration of branched tetraethers, CBT)重建该地 44.4~ 22.8 ka.BP的古温度年平均气温(mean annual air temperature, MAAT),MAAT 与频率磁化率曲线和北大西洋沉积物有孔虫 曲线对比,其变化趋势具有一致性,反映了其指标的可靠性.巫山黄土的陆源输入指数(BIT 值,bGDGTs vs. crenarchaeol)在 0.5~1.0 之间,大部分值接近于 1.0,但有几个极低值,分析为干旱事件.根据 MAAT 和 BIT 指标,巫山地区在冰期气候有剧烈 的波动,在 36.2±3.6 ka.BP、26.0±2.7 ka.BP、23.7±2.3 ka.BP 时相对比较干旱,推测可能分别对应着 H4、H3 和 H2 事件,表 明该区气候变化响应全球气候变化.

关键词:黄土;古气候;微生物醚类化合物;年平均气温;气候变化. 中图分类号:P66 **文章编号:**1000-2383(2018)11-4018-09

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# Distribution and Paleoclimate Implication of Microbial Tetraether Lipids in Wushan Loess

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**Abstract:** "Wushan loess", regarded as the southern boundary of Chinese Loess, is distributed in the Three Gorges area in the Yangtze River, which contains rich paleoclimatic information attracting attentions of many scholars in paleoclimate. In order to further reveal its causes and better understand the paleoclimatic significance contained in Wushan loess, the glycerol dialkyl glycerol tetraethers (GDGTs) extracted from Wushan loess section were investigated. Comparing the mean annual air temperature (MAAT) reconstructed based on branched GDGTs (bGDGTs) with frequency-dependent magnetic susceptibility and North Atlantic sediments, it is found that the three curves are analogous in climate evolution patterns. These similarities show that climatic changes in Wushan region respond to global climatic changes. The BIT values range from 0.5-1.0 and most BIT (bGDGTs vs. crenarchaeol) values are close to 1. The low BIT values suggest that it was very dry at 36.2 ka.BP, 30.2 ka.BP and 23.5 ka.BP. Based on MAAT and BIT values, it is found that it was relatively cold when it was very dry. It is proposed that the three periods correspond to H4, H3, H2 climate events, respectively.

Key words: loess; paleoclimate; glycerol dialkyl glycerol tetraether; mean annual air temperature; climate change.

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来自微生物细胞膜脂的甘油二烷基甘油四醚化 合物(glycerol dialkyl glycerol tetraethers, GDGTs),包括类异戊二烯GDGTs(iGDGTs)和支 链GDGTs(bGDGTs)两大类(Yang et al., 2013), 广泛存在于海洋(Lü et al., 2014)、湖泊(Günther et al., 2014)、泥炭(Coffinet et al., 2015)和土壤 (Wang et al., 2014)等沉积物中,其分布能灵敏地 响应气候环境变化,成为近年来古环境重建方面研 究的热点.

古菌能够合成含有五元环或六元环的 iGDGTs 化合物(图 1).研究者发现其大量存在于海洋 (Schouten et al., 2002)、湖泊(Buckles et al., 2014)等沉积物中,后来发现在土壤(Weijers et al., 2006b)中也有 iGDGTs 分布,其含量主要受土壤 pH(Yang et al., 2012; Dang et al., 2016)和湿度 (Wang et al., 2013)的控制.细菌 bGDGTs 一般由 2~6个甲基支链和 0~2个五元环组成(图 1),由厌 氧异养细菌生成,主要存在于土壤(Weijers et al., 2007)和泥炭(Weijers et al., 2006a)沉积物中,并 被河流带入海洋和湖泊(Herfort et al., 2006; Kim et al., 2007),另外,在湖泊和边缘海中也有自生的 bGDGTs(Tierney and Russell, 2009; Sinninghe Damsté, 2016).甲基化指数 MBT(methylation index of branched tetraethers)与环化指数 CBT(cyclization ration of branched tetraethers)与大气温度、土 壤 pH关系密切,已经成为古环境重建的重要指标 (Zhou et al., 2011; Peterse et al., 2012; Tang et al., 2017).BIT(bGDGTs vs. crenarchaeol)指标一开始用 来反映海洋、湖泊中陆源有机质的输入(Hopmans et al., 2004),近年来发现土壤 GDGTs 得到的 BIT 值能 间接反映流域水文情况,且与土壤含水率呈正相关关 系(Dirghangi et al., 2013).

黄土是晚新生代古气候研究的重要载体.前人 继利用磁化率(Hao et al., 2008; Li et al., 2015)、 粒度(Sun et al., 2008)、同位素地球化学(Wang et al., 2007; Wei et al., 2015)等方法对其进行广泛 研究之后,又把基于微生物类脂物的相关指标用于 黄土古气候环境重建(Huguet et al., 2012; Yang et al., 2014; Schreuder et al., 2016)."巫山黄土" 是指分布于长江三峡的黄土状堆积,是中国黄土的 重要组成部分,含有丰富的气候与环境变化信息,早 已引起学者关注.前人对巫山黄土的研究,主要集中 于其成因的探讨(黄臻等,2010; 李长安等,2010; 张玉芬等,2010),缺乏古环境重建方面的研究.本文 以巫山黄土剖面 GDGTs 为研究对象,分析其分布 特征,重建古温度,探讨该区域古气候环境演变特



Fig.1 The structures of GDGTs and their protonated mass to charge ratios (m/z)

征,为全球气候环境变化提供区域资料.

## 1 研究区概况和样品分析方法

#### 1.1 样品采集及定年

巫山县位于重庆市最东端,地处三峡库区腹心, 地势南北高中间低,为典型的亚热带季风湿润气候, 年均气温 18.4 ℃,年均降水量 1 041 mm,雨热同期. 样品采自巫山县客运港附近长江中游河谷的北岸, 为一个建筑工地开挖的新鲜露头,剖面高达 10 m 多,出露完整,整体以黄色或褐黄色的粉砂和砂质粘 土组成,岩性均一、无层理、未见古土壤,是理想的研 究巫山黄土的剖面(图 2).

对剖面进行系统采样,采样间隔为 5 cm,共得 样品 177 个,测试时每隔 3 个样品,即每隔 20 cm 取 样进行 GDGTs 测试;同时,测试 1 个表土 GDGTs 样品,进行温度重建对比研究,共测试 46 个黄土 GDGTs 样品.频率磁化率可以反映气候温暖湿润的 强弱和时间,为了进行对比,笔者对所有样品(177 个)进行磁化率测试,巫山黄土频率磁化率的测试工 作在中国地质大学(武汉)岩石磁学实验室完成,采 用 HKB23 型磁化率仪,其灵敏度可达 2×10<sup>-7</sup> SI. 另外,在剖面 2.00 m、4.00 m、6.00 m、8.00 m、 10.00 m处避光密封采集 5 块样品,送到中国科学院 青海盐湖研究所资源与化学重点实验室进行光释光 (OSL)测年,测年方法详见赖忠平等(Lai et al., 2007; Kang et al., 2011).由于在 8.00 m 处采得的

表 1 巫山黄土剖面 OSL 测年结果

Table 1 OSL dating results of the samples from Wushan loess section

采样深度(m)	估计光释光年龄(ka.BP)
2	$26.4 \pm 2.3$
4	$28.2 \pm 2.5$
6	$31.9 \pm 2.9$
10	44.4±4.2

样品的测年数据明显不符合实际,故将其舍去,最终的年代数据(表 1)与深度呈较强的线性关系(*R* = 0.975)(吴可等,2014),表明研究区的巫山黄土剖面沉积连续,其年代范围为 44.4~22.8 ka.BP,为晚更新世中后期沉积黄土.笔者去除风化表面后,在剖面 1 m 处开始采样.

#### 1.2 GDGTs 的萃取和分析

黄土样品经过 48 h 冷冻干燥并用玛瑙研钵研 磨过 60 目筛后,称取 80 g 左右样品和一定量的石 英砂混合均匀后装入加速溶剂萃取仪(ASE100; Dionex, USA)的萃取池中,安装好萃取池后,调好 模式的仪器自动在萃取池温度升到 100 ℃,氮气加 压至 1.0×10<sup>6</sup> Pa 后,用 DCM: MeOH(9:1, v/v) 混合溶剂萃取样品 30 min.完成萃取后将提取的脂 类转移到旋转蒸发仪浓缩至 1~2 mL,后用氮气吹 干.然后,过硅胶层析柱分离非极性组分和极性组 分,洗脱剂分别为正己烷和甲醇.极性组分用氮气吹 干后,用 DCM 重新溶解过 0.45 μm PTFE 滤膜,再 用氮吹仪吹干准备分析其中的 GDGTs.为防止人为 污染,每隔 10 个样品设 1 个空白样,实施监测.



图 2 研究区地理位图 Fig.2 Location of research region 地理位置图据百度地图(map.baidu.com)改绘

把待测样品重新溶解在 300 µL 正己烷/异丙醇 (99:1,v/v)混合溶剂中,并加入 10 µL C46 GDGT 作为内标(浓度为 0.001 157 μg/μL).类脂物 GDGTs 化合物的检测采用高效液相色谱质谱一质谱联用仪 (LC-MS),仪器型号为 Agilent 1200 HPLC,是配有 大气压化学电离源(APCI)与电喷雾(ESI)两种离子 源的 6460A 三重四级杆质谱仪,色谱柱为 Alltech Prevail 氰基柱(150 mm×2.1 mm,3 µm).GDGTs 化合物检测条件为:流动相 A 相为正己烷, B 相为 正己烷/异丙醇(体积比为9:1).洗脱梯度为0~ 5 min,90%A:10%B;5~45 min,A相比例由 90% 线性降至 82%,而后冲洗色谱柱至 B 相含量回到 10%,过程中流速为 0.2 mL/min,柱温箱恒温40 ℃. 在 APCI 源中进行化合物离子化, APCI/MS 条件如 下:雾化器压力 0.41 MPa,雾化温度 400 ℃,干燥气 (N<sub>2</sub>) 流速 6 L/min, 温度 200 ℃, 毛细管电压 3500 V, 电晕电流 5 µA(约 3200 V). 采用选择离子 扫描模式(SIM)检测 GDGTs 质子化后有特定质荷 比的离子,扫描质核比(m/z)为1302、1300、1298、 1 296, 1 292, 1 050, 1 048, 1 046, 1 036, 1 034, 1 032, 1022、1020、1018 和 744.本研究主要是对 GDGTs 化合物的定性分析,通过各离子峰面积与加入内标 面积的比值对目标化合物进行定量分析.

上机测试结果显示:样品 ws-169 的数据出现错误,将其舍去,最终得到 45 个样品数据;所有空白样均未监测出本实验所测类脂物.

### 2 结果与讨论

#### 2.1 GDGTs 分布特征

巫山黄土样品中检测出 5 种类异戊二烯类和 9 种支链类 GDGTs,古菌 iGDGTs 和细菌 bGDGTs 总 含量差别较大(图 3),分别为 31.20~3 999.40  $\mu$ g • g<sup>-1</sup>和 164.60~36 451.30  $\mu$ g • g<sup>-1</sup>.bGDGTs 占总 GDGTs 含量的 78.33%~94.29%.古菌 iGDGTs 的主 要组分为 GDGT-V和 GDGT-IV(crenarchaeol),占古 菌 iGDGTs 总量的 9.37%~71.19%.细菌 bGDGTs 中 丰度较高的组分依次为 GDGT-I、GDGT-Ib、GDGT-II、GDGT-IIb,占细菌 bGDGTs 总量的 8.52%~ 45.48%.黄土为中到弱碱性土壤,黄土中古菌 iGDGTs 的含量(相对内标 C46GDGT)比酸性土壤的含量高 (杨欢,2014),黄土中细菌 bGDGTs 的主要组分相较 于酸性土壤以 GDGT-I、GDGT-II、GDGT-III为主有 较大区别.

#### 2.2 巫山黄土 GDGTs 古环境意义

古菌 iGDGTs 和细菌 bGDGTs 所建立的古气 候环境指标已在海洋、湖泊、泥炭、热带的砖红壤和 砖黄壤中证实了其应用潜力.巫山黄土样品中检测 出了较为丰富的 GDGTs 化合物,这为利用巫山黄 土 GDGTs 重建古气候环境提供了可能性.GDGTs 化合物的 MBT 和 CBT 指标被广泛应用到古温度 的重建,目前土壤沉积物已经建立了多种全球土壤 温度校正公式(Weijers et al., 2007; Peterse et al., 2012)、中国区域校正公式(Yang et al., 2014). 由于本文采集的样品与杨欢建立的中国区域校正公 式的样品具有相同的属性,所以引用杨欢的中国区 域校正公式.笔者将探讨通过中国区域校正公式计 算的平均温度 MAAT 以及 Hopmans 等建立的陆 源输入指数 BIT (branched and isoprenoid tetraether index)(Hopmans et al., 2004)来了解巫山 黄土中蕴含的古环境信息.具体计算方法如公式 (1)、(2)、(3)、(4)(公式中罗马数字代表图1中对应 的 GDGTs 分子结构):

MBT =

$$\frac{\mathrm{I}+\mathrm{Ib}+\mathrm{Ic}}{\mathrm{I}+\mathrm{Ib}+\mathrm{Ic}+\mathrm{II}+\mathrm{IIb}+\mathrm{IIc}+\mathrm{II}+\mathrm{IIb}+\mathrm{IIc}},$$
(1)

$$CBT = -\log(\frac{I b + II b}{I + II}), \qquad (2)$$

$$MAAT = 7.5 + 16.1 \times MBT - 1.2 \times CBT, \qquad (3)$$



图 3 样品的部分基峰色谱

Fig.3 HPLC-APCI/MS base peak chromatograms of two samples in loess



图 4 巫山黄土剖面 GDGTs 各参数的变化及 bGDGTs 重建的温度与频率磁化率、北大西洋 V23-81 钻孔沉积物有孔虫曲线对比 Fig.4 Variations of GDGT proxies in Wushan loess section and comparison of bGDGT-derived MAAT with frequencydependent magnetic susceptibility and North Atlantic Core V23-81 N. Pachyderma

据 Bond et al.(1992)

$$BIT = \frac{I + II + III}{I + II + III + III}, \qquad (4)$$

所有测式及计算获得的数据如图 4.

利用公式(1)和(2)得到巫山黄土剖面中细菌的 甲基化指数 MBT 和环化指数 CBT 的变化范围分 别为:0.50~0.85,0.05~0.47.利用公式(3)计算出表 土层的温度为18.6 ℃,比现在当地记录的平均温度 18.4 ℃高,但差别不大.44.4~22.8 ka.BP, MAAT 变化范围为 15.5~18.3 ℃,平均温度约为 16.7 ℃, 比现在该区记录的温度低 1.7 ℃,这与该时段处于 冰期相比现代温度要低的事实相符.不同的纬度末 次冰期温度与现代温度的差异较大.一般来说,高纬 度地区可以相差 8~10 ℃(卢冰等,2001),低纬度地 区可能相差 2~3 ℃(姚龙奎等,2007),本研究区所 处的中纬度地区相差 4~5 ℃(李铁刚等,2006). Weijers et al.(2011)通过研究全球 8 个采样点的土 壤,发现 MBT-CBT 计算得出的温度比实际测量的 大气年均温度高 2.1 ℃.频率磁化率可反映样品中超 顺磁磁颗粒含量的多少,这些含量又反映了气候温 暖湿润的强弱和时间,温湿程度越高则频率磁化率 的值越大(刘秀铭等,1990).重建的 MAAT 曲线与 频率磁化率相比较(图 3),两条曲线总体上的变化 趋势基本一致,虽然重建巫山地区的古温度 MAAT 曲线绝对温度上偏高,但其相对变化趋势是可信的.

图 4 为巫山黄土重建的古温度和北大西洋钻 V23-81 有孔虫含量对比曲线(Bond *et al.*,1992),有相似 的气候变化,在该时期,温度有较大的波动,尤其在 6.6 m、4.8 m 和 1.6 m 处气温有大幅度降低.

本文由公式(4)得到的 BIT 值大部分接近 1 (图 4),一般在 0.95~1.00 范围内波动,但受当时具 体环境影响,部分样品的 BIT 值有所波动,分别在 6.6 m、4.8 m 和 1.6 m 处出现极低值,对应地 BIT 值 分别降到 0.50、0.92、0.80,反映了这些时期出现了 干旱事件.一般 BIT 指数的范围为 0~1,在土壤中, 支链 GDGTs 含量很高,BIT 指数接近于 1;而在远 洋沉积物中几乎没有支链 GDGTs,BIT 指数则接近 于 0.尽管土壤中 BIT 接近于 1 的那些值不能反映湿 润气候,但 BIT 的那些相对极低值可以反映干旱事 件.前人研究表明:中到弱碱性条件下,越干旱厌氧细 菌所产生的 bGDGTs 越少,好氧所产生的古菌越多, 从而导致 BIT 值变小(Xie et al., 2013; 杨欢,2014).

根据图 4,在 6.6 m、4.8 m 和 1.6 m 处 BIT 为极 低值,同时 MAAT 也是低值,反映了干旱事件,极 干对应着寒冷.根据测年插值计算,误差取测试中间 值的平均,这 3 个低值对应的时间段分别为 36.2± 3.6 ka.BP、26.0±2.7 ka.BP 和 23.7±2.3 ka.BP,这 些年龄范围在国际公认的 H4、H3、H2 年龄范围 内,笔者推测此3次事件对应于北大西洋沉积物所 指示的 H4、H3 和 H2 事件(Bond et al., 1992).从 数据上分析认为 H4 最干旱, H2 次之, H3 最末, 可 能 H4 事件正好对应着 D/O 旋回的寒冷期, 而 H3、 H2事件对应着 D/O 旋回的间冰阶期(王绍武, 2011).结合获得的 MAAT 和 BIT 分析,在巫山地 区,极端干旱时期对应着温度的降低,可能是中北大 西洋冰漂碎屑事件(H事件)(Heinrich, 1988; Bond et al., 1992), 通过海一气系统的快速重组, 西风带和蒙古冷高压影响东亚季风气候系统(An, 2000),导致全球气候突发性降温干旱.图 3 中巫山 黄土曲线所反映的千年尺度气候突变与北大西洋 V23-81 钻孔沉积物记录的 Heinrich 事件对应较 好,说明研究时期内巫山短暂的气候回返事件具有 全球性.但巫山黄土指示的气候没有较明显的周期 性,与格陵兰冰芯记录有一定的差异性,可能是受巫 山复杂地理环境的影响以致呈现一定的区域性.

### 3 结论

(1)巫山黄土检测出了丰富的微生物类脂物,其 中细菌 bGDGTs 含量明显高于古菌 iGDGTs 含量, 表现出黄土中到弱碱性土壤的 GDGTs 分布特征.

(2)利用 MBT 和 CBT 重建的巫山黄土末次冰 期间的 MAAT 值虽然比实际偏高,但变化趋势与 其他指标一致,可反演气温的相对变化,MAAT 值 波动较大,反映了冰期气候的不稳定性.巫山黄土的 BIT 指数大部分接近于 1,但局部由于极端气候事 件的影响有降低,指示干旱事件.根据 MAAT 和 BIT 指标,巫山地区在约 36.2±3.6 ka.BP、26.0± 2.7 ka.BP和 23.7±2.3 ka.BP 时对应着干冷,推测可 能分别对应着 H4、H3 和 H2 事件.

(3) 巫山短暂的气候回返事件具有全球性,但又 与北大西洋记录有一定的差异性,可能是受巫山复 杂地理环境的影响致使呈现一定的区域性.

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