论文

生物有机指标在气候和环境变化研究中的应用进展专题

青藏高原东北部青海湖距今 12 ka 以来的四醚指标 BIT 记录: 对古环境重建的启示

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摘要 早期的研究认为湖泊沉积物中的支链 GDGTs(bGDGTs)主要来自陆地土壤细菌, 而奇古菌醇由水生奇古菌产生, 因此湖泊中表示 bGDGTs 与奇古菌醇比值的 BIT 指标可用于衡量陆源有机质的输入程度. 然而越来越多的研究表明, 湖泊环境中普遍存在着自生的 bGDGTs, 这样, 湖泊沉积物中的 BIT 指标可能不能用来指示陆源物质输入. 尽管如此, 最近对非洲 Challa 湖的研究表明 BIT 仍可作为一个有效的古水文指标, 但是在不同的湖泊中 BIT 对古水文变化的响应可能有所不同. 本文调查了距今 12 ka 以来青海湖沉积物中的 BIT 及其相关 GDGTs 含量的变化. 结合已发表的表层样品数据发现, 青海湖沉积物中 BIT 的变化主要反映了奇古菌醇含量的变化, 而不是细菌 bGDGTs 含量的变化. 在青海湖中, 由于奇古菌醇含量与水深正相关, 因此 BIT 与水深负相关. 这与 Challa 湖 BIT 与湖面或降雨正相关的情况相反, 表明不同湖泊中 BIT 对水文变化的响应有所差异. 因而在应用 BIT 重建特定湖泊古水文变化时, 需要先确定控制其沉积物 BIT 值的主要因素.

关键词 青海湖, BIT, 湖水深度, 降雨量, 古水文

1 引言

甘油二烷基甘油四醚类化合物(GDGTs)主要包括类异戊二烯 GDGTs(iGDGTs)和支链 GDGTs

(bGDGTs). 这些化合物比较容易分析, 且不同系列 GDGTs 的分布对不同的环境参数敏感(Schouten等, 2007a; Castañeda和Schouten, 2011; 杨欢等, 2012; Schouten等, 2013), 因而GDGTs在古环境研究中应用

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比较广泛. iGDGTs来源于生命三域之一的古菌(Woese等, 1990)的细胞膜脂(De Rosa和Gambacorta, 1988; Schouten等, 2013). 在iGDGTs中, 奇古菌醇被认为是奇古菌特有的生物标志化合物(Sinninghe Damsté等, 2002a; Brochier-Armanet等, 2008; Pitcher等, 2010), 它们在海洋和一些较大的湖泊中含量很高(如, Schouten等, 2002; Sinninghe Damsté等, 2002b; Turich等, 2007; Kim等, 2008; Powers等, 2010; Tierney等, 2010). bGDGTs是另一类主要的GDGTs, 它们可能是某(些)厌氧(Pancost等, 2003; Oppermann等, 2010; Weijers等, 2010)细菌(Weijers等, 2006; Sinninghe Damsté等, 2011)的生物膜脂成分, 早期的研究发现bGDGTs主要存在于土壤和泥炭中(Hopmans等, 2004). 因此Hopmans等(2004)定义了一个表示三种主要bGDGTs与奇古菌醇相对含量比值的BIT(Branched and Isoprenoid Tetraether)指标, 并用该指标来衡量水相环境中陆源有机质与水体有机质的相对输入程度. 目前, BIT指标已被广泛应用于钻孔和表层沉积物以示踪土壤有机质对湖泊和海洋环境的贡献(Herfort等, 2006; Ménot等, 2006; Schouten等, 2007b; Seki等, 2009; Weijers等, 2009; Verschuren等, 2009; Sun D等, 2011).

在湖泊中, 尽管BIT具有示踪陆源土壤有机质输入(Hopmans等, 2004; Blaga等, 2009; Verschuren等, 2009)或土壤iGDGTs输入(Powers等, 2010)的前景, 越来越多的证据表明湖泊里存在着大量的水体或沉积物中自生的bGDGTs. 首先, 不同地区湖泊的研究均显示沉积物中bGDGTs浓度比土壤中的高得多, 且bGDGTs分布与周边土壤中的显著不同(Sinninghe Damsté等, 2009; Tierney和Russell, 2009; Tierney等, 2010; Sun Q等, 2011; Loomis等, 2011, 2014; Peterse等, 2014; Buckles等, 2014a, 2014b; Naeher等, 2014). 其次, 沉积物中有大量以完整极性膜脂(IPLs)形式存在的bGDGTs而土壤中的IPL bGDGTs很少(Tierney等, 2012; Wang等, 2012; Buckles等, 2014b; Huguet等, 2015). 再次, 最近Weber等(2015)在瑞士的一个高山湖泊的沉积物中发现了一种新的bGDGT, 而湖区周边土壤中没有检测到这种bGDGT; 他们还发现沉积物中bGDGTs裂解产物的稳定碳同位素值比周边土壤中的要偏负. 另外, 在其他水相环境中, 如河流和河口地区(Zhang等, 2011; Zhu等, 2011; Yang等, 2013;

Zell等, 2013; De Jonge等, 2014; Zhou等, 2014)以及边缘海和远洋环境里(Peterse等, 2009; Fietz等, 2012; Hu等, 2012; Liu等, 2014; Weijers等, 2014; Dong等, 2015)也可能存在着自生的bGDGTs. 这些水相环境中自生的bGDGTs势必会妨碍应用BIT指标重建陆源土壤有机质的输入(Schouten等, 2013). 因此, 沉积物中BIT指标的地球化学意义还需要更深入的研究.

Challa湖是非洲的一个火山口小湖, 前人对该湖中的BIT指标进行了比较系统的研究. Sinninghe Damsté等(2009)观测到Challa湖的bGDGTs主要沉降过程发生在强降雨季节, 因此他们认为该湖沉积物中的bGDGTs可能主要来自地表径流带来的流域内的土壤, 尽管不能排除湖泊和地下水中自生bGDGTs的贡献. 根据这个观测结果, Verschuren等(2009)利用Challa湖钻孔沉积物重建了距今25 ka以来的区域降雨变化, 他们发现BIT记录与根据地震反射地层结果推断的湖面波动历史(Moernaut等, 2010)符合的很好. 然而, 后来Sinninghe Damsté等(2012)注意到该钻孔中BIT的变化主要取决于奇古菌醇含量的变化, 而非bGDGTs含量的变化. 因此, 他们推测BIT与降雨(或湖面)的同步变化可能反映了一些与降雨有关的参数(如: 湖面、风力和土壤营养物质输入等)对湖泊养分循环的影响. 随后, 根据一些现代过程的证据, Buckles等(2014b)清楚的表明Challa湖中大部分bGDGTs是自生的. 为进一步探究该湖中BIT与降雨相关联的确切机制, Buckles等(2014b, 2015)调查了表层沉积物、距今2200年来的钻孔沉积物以及沉积物捕获器按月采集的样品中的GDGTs分布. 他们的结果表明间歇性的强降雨带来的土壤养分输入可能会改变湖中浮游生物和微生物的群落结构, 进而抑制奇古菌的生长, 最终导致较高的BIT值(Buckles等, 2015). 这一系列的研究显示, 在年代际或者更长的时间尺度上, Challa湖沉积物的BIT可作为一个有效的降水指标. 但Buckles等(2015)同时指出在其他地区应用BIT指标时需要谨慎, 因为在不同地区BIT对降雨的响应可能是不同的. 因此, 仍需进一步探究其他湖泊中BIT对区域水文变化的响应机制.

青海湖是中亚地区最大的内陆高原湖泊(Fu等, 2013). 在本研究中, 我们将调查距今12 ka以来的该湖QH-2011钻孔沉积物中的BIT及其相关GDGTs含量的变化. 我们希望通过研究该段钻孔沉积物中BIT指

标对与其他指标反演的湖面波动(Wang等, 2014)的响应, 以及表层沉积物(数据来自Wang等, 2012)中BIT随原位水深变化, 揭示这一地区湖泊沉积物中BIT指标对区域水文变化的响应机制。

2 材料和方法

2.1 研究区域和样品

青海湖属碱性咸水湖, 其盐度约为14~16 g/L(Liu W等, 2011), pH约为8.8~9.3(Xu等, 2010)。该湖被大通山、日月山和青海南山包围(Liu W等, 2011), 湖面面积约为4260 km², 流域面积超过29660 km², 湖水体积约为7.16×10¹⁰ m³(Jin等, 2010)。湖面海拔约为3194 m, 湖水最深处约27 m(Xiao等, 2012)。青海湖地区地处青藏高原的东北缘, 具有寒冷半干旱的气候特征。区域年平均温度约为1.2℃(1951~2005年, Jin等, 2010), 年平均降雨量约为373 mm, 其中超过65%的降水发生在夏季(6~8月), 表现出显著的季风降水的季节性特征(An等, 2012), 但是潜在蒸发量远超过降雨量(孙大鹏等, 1991)。每年青海湖有3~4个月的冰封期(12月至次年3月), 结冰最厚可达约0.8 m(Colman等, 2007)。青海湖湖水在夏季会出现分层(温跃层约在10~15 m处), 上层水温12~15℃, 下层水温约为6℃(Lister等, 1991; Williams, 1991)。主要的入湖河流有布哈河、沙流河、泉吉河、哈尔盖河以及黑马河, 它们位于湖的北部和西北部(Jin等, 2010)。

QH-2011钻孔于2011年8月采自青海湖东南湖盆水深约24 m处, 钻孔全长5.8 m。整个钻孔共测了6个AMS¹⁴C年龄。¹⁴C年龄扣除碳库效应后进行日历年龄校正(IntCal13, Reimer等, 2013), 然后与相邻钻孔的6个日历年龄(Wang等, 2011)结合, 得到分段的深度年代模式(Wang等, 2014)。本文分析了QH-2011钻孔上部4.35 m全新世以来的145个样品的GDGTs组分, 其中12个样品由于bGDGTs色谱图受到未知化合物的影响没有给出BIT值。样品的平均年代分辨率约为100年。

2.2 脂类分析

QH-2011钻孔GDGTs的提取和测量方法在Wang等(2014)中已报道。简言之, 1~5 g沉积物样品分别用甲醇、甲醇/二氯甲烷(1:1, 体积比), 二氯甲烷、甲醇

/二氯甲烷(1:1, 体积比)和甲醇超声振荡抽提。随后向5次所得的总抽提液中加入一定量的C₄₆ GDGT内标(Huguet等, 2006)并在氮气下吹干。吹干后的样品用99:1(体积比)的正己烷/异丙醇重新溶解后用0.45 μm的聚四氟乙烯滤膜过滤, 然后用高效液相色谱-大气压化学电离-质谱(HPLC-APCI-MS)检测GDGTs(Zhang等, 2012)。色谱型号为Agilent 1200 HPLC, 质谱型号为QQQ 6460 MS。扫描模式为选择离子扫描(SIM)。通过对各GDGTs的[M+1]⁺离子峰面积积分并对比其与C₄₆内标[M+1]⁺离子峰面积的比值, 可以对样品中GDGTs的含量进行定量。这里假设各GDGTs与C₄₆内标具有相同的仪器响应因子。

BIT指标按照Hopmans等(2004)的公式计算:

$$\text{BIT} = \frac{\text{I} + \text{II} + \text{III}}{\text{I} + \text{II} + \text{III} + \text{crenarchaeol}} \quad (1)$$

3 结果

过去12000年来青海湖的GDGTs组成发生了显著变化。我们之前的研究(Wang等, 2014, 2015)报道过QH-2011钻孔的部分iGDGTs数据。本文中我们新报道了奇古菌醇含量、bGDGTs含量以及BIT指标的变化情况。

沉积物中的奇古菌醇浓度为1.9~535.9 ng g⁻¹(图1(a)), 变化范围约有3个数量级。除了8.4 ka BP时奇古菌醇浓度较高, 早全新世时奇古菌醇浓度基本低于10 ng g⁻¹。中全新世奇古菌醇浓度逐渐上升并伴有百年-千年尺度的波动。晚全新世奇古菌醇浓度普遍高于整个全新世奇古菌醇浓度的平均值(140.2 ng g⁻¹)。奇古菌醇浓度最大值出现在约4.8 ka BP。

总bGDGTs浓度的变化范围为122.8~751.6 ng g⁻¹, BIT相关的三种主要bGDGTs(Ia+IIa+IIIa)浓度变化范围为100.0~713.0 ng g⁻¹(图1(b))。总bGDGTs浓度和(Ia+IIa+IIIa)浓度平均值分别为268.6和221.2 ng g⁻¹, 二者均比奇古菌醇浓度平均值要高。但是整个全新世bGDGTs浓度变化范围只有一个数量级。

BIT指标的变化范围为0.22~0.99 (平均值0.61, 图1(c)), 其变化趋势与奇古菌醇浓度的变化趋势相反。总的来说, 早全新世BIT接近于1(除了约8.4 kaBP时), 随后BIT波动下降至晚全新世的低值期(晚全新世BIT平均值为0.43)。整个记录中的BIT最小值

图1 青海湖QH-2011钻孔全新世以来GDGTs含量和BIT的变化

(a) 奇古菌醇含量; (b) bGDGTs含量; (c) BIT; (b)中黑点表示总bGDGTs, 灰点表示三种主要bGDGTs(Ia+IIa+IIIa). 水平虚线指示各参数平均值

也出现在约4.8 ka BP.

4 讨论

4.1 青海湖全新世湖面演化历史

在过去的几十年里, 尽管人们已经做了广泛的研究(如, 张彭熹等, 1989; Lister等, 1991; Yu, 2005; Ji等, 2005; Shen等, 2005; Colman等, 2007; Liu X等, 2011; An等, 2012; Li和Liu, 2014; Liu等, 2015), 但有关全新世青海湖湖面波动的连续记录还比较少. 根据地震波反射数据、碳酸盐、沉水植物种子化石、沉积构造和特征(Kelts等, 1989; Lister等, 1991; Yu, 2005)以及介形虫壳体Sr/Ca值推断的盐度(张彭熹等, 1994)等, 前人对20世纪80年代所采集钻孔的研究结果表明, 该湖在早全新世较浅而晚全新世较深. 最近, 对青海湖南盆(1F孔, 采于2005年)和东南湖盆(QH-2011)两个钻孔的高分辨率的有机地球化学水深指标的研究进一步清楚的显示: 青海湖在早全新世湖水较浅, 中全新世湖面逐渐扩张并伴随着几百年

尺度的波动, 晚全新世湖面比较稳定, 湖水一直较深(Liu等, 2013; Wang等, 2014). 青海湖全新世湖面波动历史与青藏高原东北部(Mischke等, 2008; Herzs Schuh等, 2009)以及中国西北干旱/半干旱地区(Herzs Schuh, 2006; Chen等, 2008)的一些其他湖泊记录相符. 青海湖早全新世的低湖面可能反映了干旱/半干旱地区夏季温度决定的蒸发量变化对区域有效湿度具有重要作用(Yu, 2005; He等, 2013; Liu等, 2013; Wang等, 2014; Liu等, 2015).

4.2 青海湖中的自生bGDGTs

我们之前对表层沉积物和周边土壤的研究结果显示, 青海湖中至少有部分bGDGTs是自生的(Wang等, 2012). 这主要基于两个证据: 首先, 表层沉积物中bGDGTs的分布(例如MBT和CBT指标)与周边土壤有显著不同. 这一现象与大部分湖泊的观测结果是一致的(Sinninghe Damsté等, 2009; Tierney和Russell, 2009, 2010; Sun Q等, 2011; Loomis等, 2011, 2014; Peterse等, 2014; Buckles等, 2014a, 2014b; Naether等,

2014), 很少有例外(如, Niemann等, 2012). 其次, 青海湖表层沉积物中IPL bGDGTs(即带有极性头基团的bGDGTs)的相对含量比周边土壤中的要高. 最近一些其他湖泊的研究结果也表明湖泊沉积物的IPL bGDGTs含量较高(Tierney等, 2012; Buckles等, 2014b, Huguet等, 2015). 尽管IPL GDGTs不能完全表示活体生物量(Schouten等, 2010), 但是如果认为湖泊中的bGDGTs只来自于周边土壤, 那么沉积物中相对较高含量的IPL bGDGTs是无法解释的(Buckles等, 2014b).

此外, 对比青海湖表层沉积物和周边土壤的BIT值, 我们发现虽然离岸沉积物的BIT值比周边土壤的BIT值要高, 但水深<1 m的近岸沉积物的BIT值通常比土壤的BIT值要高(图2(a)). 因此, 即使近岸沉积物的奇古菌醇完全来自土壤, 也必定存在湖泊自生的bGDGTs, 它们导致了这些沉积物中的BIT值增高.

总之, 尽管目前尚不能排除陆源土壤输入对青海湖bGDGTs的贡献, 然而青海湖中确实存在着自生

的bGDGTs. 这会妨碍在该湖中应用BIT指标来示踪土壤有机质的输入. 因此, 有必要进一步研究青海湖中影响BIT的环境因素, 而不能简单地将其当作陆源有机质输入的指示(Wang等, 2012).

4.3 奇古菌醇对 BIT 指标的控制

在QH-2011钻孔中, 表示bGDGTs与奇古菌醇相对含量的BIT指标变化显著, 而且BIT的变化与奇古菌醇含量的变化非常一致(图1). 相比之下, bGDGTs的含量比较稳定(图1). 我们也注意到青海湖表层沉积物中奇古菌醇含量随水深的增加而升高, 但bGDGT含量随水深变化不明显(图2(b)), 这导致BIT随水深增加而降低(图2(a)). 因此, 在青海湖沉积物中BIT的变化似乎主要受控于奇古菌醇含量的变化, 而bGDGTs则类似背景的信号. 我们将QH-2011钻孔的这两类GDGTs分别与BIT做相关性分析时, 奇古菌醇含量对BIT的控制作用更加明显. 如图3所示, 奇古菌醇含量与BIT有很强的负相关关系($R^2=0.85$), 而bGDGTs含量与BIT的关系相对较弱($R^2=0.57$). BIT与奇古菌醇含量的显著的相关关系在40余个非洲湖泊表层沉积物中(Tierney等, 2010), 以及Challa湖(Sinninghe Damsté等, 2012; Buckles等, 2015)、Baikal湖(Fietz等, 2011)和一些海洋环境(Fietz等, 2011; Smith等, 2012)的钻孔沉积物中也都有报道.

4.4 青海湖中水深对 BIT 的作用

由于青海湖中BIT的变化主要取决于奇古菌醇的产量, 而后者与水深密切相关(Wang等, 2014), 我们推断该湖中BIT的变化也受湖水深度变化的显著影

图2 青海湖中BIT与水深的关系以及GDGTs含量与水深的关系

数据来自 Wang 等(2012). 为便于对比, (a)中也给出了周边土壤的BIT值

图3 QH-2011 钻孔上部 4.35 m 样品 BIT 与 GDGTs 含量的关系

响. 事实上, 对钻孔沉积物来说, QH-2011孔的BIT记录与QH-2011钻孔的奇古菌醇相对含量(%cren)以及1F孔和QH-2011钻孔的总有机碳同位素($\delta^{13}\text{C}$)重建的湖面变化大体一致(图4, Liu等, 2013; Wang等, 2014). 其中早全新世湖面较低时BIT值较大, 而晚全新世湖面较高时BIT值较小. 特别是对于同一钻孔(QH-2011), BIT的波动与%cren和 $\delta^{13}\text{C}$ 记录基本吻合, 尤其是在早全新世和中全新世时期(Wang等, 2014). 我们也注意到, 对青海湖表层沉积物来说(Wang等, 2012) BIT与水深同样显著相关(图2(a)). 另外, 在具有不同理化参数的41个非洲湖泊(Tierney等, 2010)和82个全球分布的湖泊中(Blaga等, 2010), BIT均与水深显著负相关(图5). 这些结果一致表明, 对于青海湖和很多其他湖泊来说, 水深对沉积物中的BIT值都具有重要的控制作用.

4.5 与 Challa 湖的对比: 对 BIT 指标古环境应用的启示

BIT与水深的相关性表明其仍可作为古水文重建的指标. 但是, 在Challa湖中BIT与湖面或降雨量

正相关(Verschuren等, 2009; Buckles等, 2015), 而在青海湖中BIT与水深负相关. 这种差异可能是由于两种类型湖泊中控制奇古菌(奇古菌醇的母源)生物量的机制是不同的.

最近的观测结果表明奇古菌(以及奇古菌醇)在水体的温跃层/氧跃层丰度最高(Pouliot等, 2009; Llirós等, 2010; Auguet等, 2012; Schouten等, 2012; Woltering等, 2012; Buckles等, 2013). 因此在青海湖中, 奇古菌醇含量与水深的正相关关系可能反映了水生奇古菌喜欢生活在湖水中较深的(次表层)区域(Wang等, 2014). 另外一个可能的解释是, 对于一个特定湖泊(如青海湖)的特定区域, 水深一般和离岸距离相关, 而离岸距离又会影响到地表径流带来的营养盐和陆源有机质输入量, 进而影响湖泊的初级生产量. 一般认为, 具有氨氧化功能的奇古菌是自养的(Pester等, 2011), 且适应于低氨氮浓度的环境(Martens-Habben等, 2009; Pester等, 2011). 但在浅水区(自生的和外源的)有机质降解会产生较高含量的铵盐, 致使奇古菌在与氨氧化细菌竞争时处于劣势(Di等, 2009; Buckles等, 2015). 因此, 在陆源有机质

图4 青海湖全新世以来 BIT 记录与各湖面指标变化对比

(a) 总有机碳同位素 $\delta^{13}\text{C}$ 记录(黑: Liu 等, 2013; 灰: Wang 等, 2014); (b) 奇古菌醇相对含量 %cren 记录(Wang 等, 2014); (c) BIT 记录

图5 82个全球分布的湖泊以及41个非洲湖泊表层沉积物中BIT与水深的关系

灰色数据来自 Blaga 等(2010), 黑色数据来自 Tierney 等(2010). 考虑到不同实验室(以及不同的分析条件)测量同一样品可能会得出不同的 BIT 值, 两组数据未合并到一起

和营养盐输入较多的近岸浅水区域, 奇古菌的生长可能会受到抑制. 这样, 对于寡营养的青海湖中的一个特定的点位, 湖泊收缩既会导致湖水变浅也可能增加营养物质的供应, 二者均会抑制奇古菌的生长.

Challa湖是赤道东非一个火山口湖, 其面积很小, 只有4.2 km², 但是水深很深(最深处约95 m), 具有典型火山口湖盆所具有的水下陡坡(Moernaut等, 2010; Sinninghe Damsté等, 2011). 该湖的湖面变化主要受当地降雨和径流的影响(Moernaut等, 2010), 高湖面一般与多的降雨量和暴雨次数有关. 另一方面, 对于这样一个水下边坡很陡的小湖, 湖中一个特定点位与湖岸的距离随湖面的升高变化很小, 因而, 陆源营养物质和有机质的供应随湖面升高和降雨增加而增加, 致使奇古菌的生长受到抑制(Buckles等, 2015). 另外, 由于Challa湖湖水很深, 水深可能不再是限制奇古菌生长的一个主要因素. Challa湖的这种情形与青海湖的非常不同. 对青海湖来说, 区域有效湿度增加会引起湖泊的扩张, 尽管径流量增大可能会使带入湖中的陆源有机质和营养盐增加, 但对于一个特定点位, 离岸距离也会增大, 致使传输到该位置的陆源有机质和营养盐可能反而减少, 这有利于寡营养的奇古菌的生长. 另外, 青海湖的扩张也伴随着水深的增加, 这也有利于水生奇古菌的繁盛. 因此, 在Challa湖中沉积物BIT通常与区域湿度(或湖面)正相关而在青海湖中恰恰相反.

5 结论

青海湖沉积物中BIT的变化主要受奇古菌醇含量控制. 由于该湖中奇古菌醇含量与水深正相关, 所以BIT与水深负相关. 这种情形似乎与Challa湖(一个赤道东非火山口小湖)相反, 因为在Challa湖中沉积物BIT值与湖面和降雨量是正相关的(Verschuren等, 2009; Buckles等, 2015). 二者之中BIT与水文条件关系的差异可能是由于不同类型湖泊中影响奇古菌的湖泊条件不同. 因而, 在不同湖泊系统里BIT对区域水文变化的响应可能是有差异的. 但是我们注意到, 对理化参数跨度很大的区域和全球湖泊群(Blaga等, 2010; Tierney等, 2010), 表层沉积物中的BIT值与水深均呈负相关关系, 这表明对大部分湖泊来说沉积物BIT值与水深是负相关的. 然而, 对于一个特定的湖泊, 在应用BIT重建其古水文变化之前, 仍有必要进行详细的调查以确定BIT是如何响应区域水文变化的.

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