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网湖沉积物正构烷烃分布特征及其记录的环境变化

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摘要: 通过对网湖沉积岩芯中正构烷烃含量和组成特征的分析, 探讨了网湖近百年来湖泊环境变化。结果表明, 网湖沉积岩芯中正构烷烃的碳数范围在 $n\text{-C}_{14} \sim n\text{-C}_{33}$ 之间, 其中以高碳数组分为主, 并具有明显的奇偶优势, 反映了沉积物有机质以大型水生植物和陆生植物贡献为主, 较低的 $2n\text{-C}_{31}/(n\text{-C}_{27} + n\text{-C}_{29})$ 比值指示陆源输入中以木本植物输入为主。根据正构烷烃参数指示的沉积物有机质来源变化特征, 近百年来网湖水环境变化具有如下 3 个阶段: 20 世纪 50 年代以前, 网湖与长江水体交换频繁, 湖泊水体处于低营养环境状态, 沉积物正构烷烃高/低分子量正构烷烃比值 (H/L) 和陆/水生类脂物比值 (TAR) 较高, 沉积物有机质主要来源于陆生植物和大型水生植物, 湖泊浮游藻类贡献少; 20 世纪 50 ~ 80 年代, H/L 和 TAR 值明显下降, 中、短链正构烷烃的比例略有升高, 表明陆源植被对沉积物有机质的贡献降低, 水生植物和浮游藻类贡献的有机质增加, 但较低的 $2n\text{-C}_{17}/(n\text{-C}_{23} + n\text{-C}_{25})$ 值表明浮游藻类有机质较低, 此时湖泊水体较为稳定, 湖泊受长江水位影响减小, 湖泊营养水平有所升高; 1980s 以来, 总体上湖泊受流域人类活动影响明显, 湖泊水体营养水平升高, 沉积物正构烷烃表现为 H/L 和 TAR 值升高后下降, 正构烷烃总量和 $2n\text{-C}_{17}/(n\text{-C}_{23} + n\text{-C}_{25})$ 比值显著升高, 2000 年后尤其明显, 表明湖泊沉积物有机质输入增加, 其中湖泊浮游藻类贡献明显增加。

关键词: 长江中游; 网湖; 沉积岩芯; 正构烷烃; 有机质来源; 环境变化

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Distribution of *n*-alkanes from Lake Wanghu Sediments in Relation to Environmental Changes

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Abstract: Concentrations and distribution of *n*-alkanes ($n\text{-C}_{14}$ to $n\text{-C}_{33}$) in Lake Wanghu sediment core were analyzed to investigate the environmental changes in the lake during the past ~ 100 years. Relatively higher concentrations of mid- and long-chain *n*-alkanes with a strong odd-over-even carbon number predominance indicated organic matter contributions dominated by aquatic macrophytes and terrestrial plants. The lower values of $2n\text{-C}_{31}/(n\text{-C}_{27} + n\text{-C}_{29})$ highlighted a type of landscape dominated by woody plants. The environmental changes that have been happening over the past 100 years in Lake Wanghu can be divided into three stages, based on the organic matter sources inferred from the changes in *n*-alkane parameters in the sediments. Before the 1950s, higher values of terrigenous/aquatic ratio (TAR) and high-/low-molecular weight *n*-alkanes (H/L) indicated vascular, plant-derived sediments deposited during a low-nutrient, clear-water phase with frequent water exchange between the lake and the Yangtze River. Between the 1950s and 1980s, decrease in values of H/L and TAR , and increase in the proportion of mid- and short-chain *n*-alkanes indicated a shift towards mid- and shorter-chain components likely associated with the increasing contribution of macrophytes and algae. Lower $2n\text{-C}_{17}/(n\text{-C}_{23} + n\text{-C}_{25})$ values indicated a relatively low algal abundance. In that stage, the lake trophic state increased slightly with the decreasing influence of the Yangtze River flow. After the 1980s, H/L and TAR values increased and then decreased, total *n*-alkane concentrations and $2n\text{-C}_{17}/(n\text{-C}_{23} + n\text{-C}_{25})$ increased significantly, especially after 2000s, suggesting the main contributor to high organic matter input most likely shifted to algae because of the eutrophication caused by anthropogenic activities.

Key words: middle reach of Yangtze River; Lake Wanghu; sediment core; *n*-alkane; organic matter sources; environmental changes

长江中下游是我国淡水浅水湖泊分布最集中的地区, 随着近几十年来区域经济快速发展, 区内湖泊水体出现富营养化和污染等问题^[1]。正构烷烃是湖泊沉积物有机质的常见组分, 作为一种生物标志物, 继承了原生物的信息。由于生物与环境之间存在密切的关联, 因此, 可以利用正构烷烃研究沉积物有机质生物源及其环境特征^[2]。

正构烷烃对气候、环境变化较为敏感, 且难被

降解, 易于在沉积物中稳定保存^[3]。另外, 不同生物来源的正构烷烃分布特征和相对含量差别较大^[4~10], 甚至在湖泊演化的不同阶段, 正构烷烃的分布模式也会呈现出规律性的变化^[11~15]。因此, 湖

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泊沉积物中的正构烷烃指标能够较为准确地反映不同生物来源有机质的相对含量变化,进而可用于推测历史时期的湖泊环境变化。目前,湖泊沉积物正构烷烃已较广泛应用于古植被、古生态和古环境等方面的研究,并取得了较好的进展^[16~19]。

迄今为止,长江中下游湖泊如太湖、巢湖、鄱阳湖等沉积物中的正构烷烃已有研究^[20~22],而对位于长江中游网湖的研究大多集中在气候变化、元素分析和洪水事件等方面^[23~26],缺少湖泊沉积物正构烷烃方面研究。本文以网湖的沉积岩芯为载体,在²¹⁰Pb、¹³⁷Cs精确定年的基础上,测定正构烷烃含量,研究其组成和相关参数的变化特征,探讨近百年来网湖环境变化,以期为湖泊水体污染控制和制定相应的对策提供理论依据。

1 材料与方法

1.1 研究区概况和样品采集

网湖位于湖北省阳新县(图1),濒临长江,西、南面为幕阜山脉北延的丘陵,东、北面为长江河漫滩平原。湖泊面积为42.3 km²,最大水深为5.4 m,平均水深为3.7 m。网湖流域的气候属亚热带季风气候,年平均气温为15.9℃,降水量为1385 mm。该地区地带性植被类型属亚热带常绿阔叶落叶混交林,亚热带针叶林占一定优势^[26, 27]。阳新县以农业为主,境内矿产资源丰富。在20世纪60年代富池口建闸以前,网湖除富水等流域径流和湖面降水补给外,受长江影响较大,每到汛期,长江水倒灌,江水携带的泥沙在湖中沉积^[23~25]。1967年富池大坝建成,阻断了网湖与长江的天然联系,使得进出网湖的湖水受人工闸口控制。

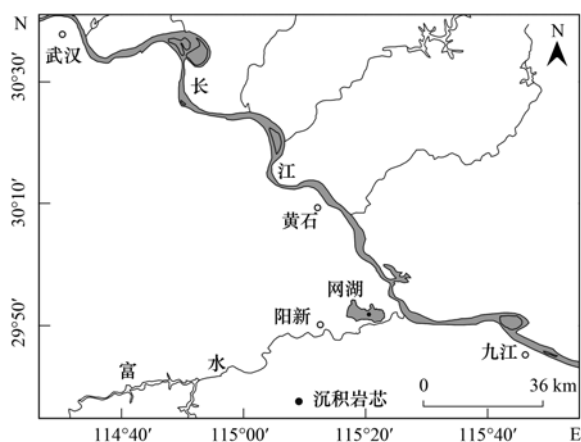


图1 网湖及其沉积岩芯的采样点示意

Fig. 1 Location of Lake Wanghu and sediment core sampling site

2015年11月利用重力采样器在网湖湖心

(29°51'49"N, 115°20'13"E)采集1根85 cm的短柱沉积岩芯,以1 cm间隔分样,样品用锡箔纸包好后密封于塑料袋内带回实验室,并于-20℃条件下冷藏以备分析。

1.2 正构烷烃提取和仪器分析

样品分析前经冷冻干燥,研磨过100目筛。称取3 g沉积物样品,加入0.2 mL已知浓度的内标(氘代正二十四烷)后,用二氯甲烷:甲醇(9:1,体积比)混合溶剂索式抽提72 h,过滤分离萃取液。旋转蒸发浓缩后转移到圆底瓶中,氮气吹干至恒重,得到湖泊沉积物中有机质的游离态部分。之后通过硅胶层析柱进行组分分离,用80 mL正己烷淋洗填充柱,得到正构烷烃组分。将洗脱后的正构烷烃组分溶液旋转蒸发浓缩、定容后进行GC/MS测定。

沉积物中正构烷烃组分分析由Agilent 7890A型气相色谱与Agilent 5975型质谱联用仪(gas chromatography-mass spectrometry, GC-MS)完成,离子源为电子轰击源(70 eV),色谱柱为DB-5MS(30 m×0.25 mm×0.25 μm)。实验中,无分流进样1 μL,进样口温度为250℃,升温程序为:初始温度80℃,保留2 min;以3℃·min⁻¹升至300℃,保持30 min。载气为高纯氦气,流速为1 mL·min⁻¹。根据相对保留时间、特征碎片离子等对化合物作定性分析,以内标作定量分析。

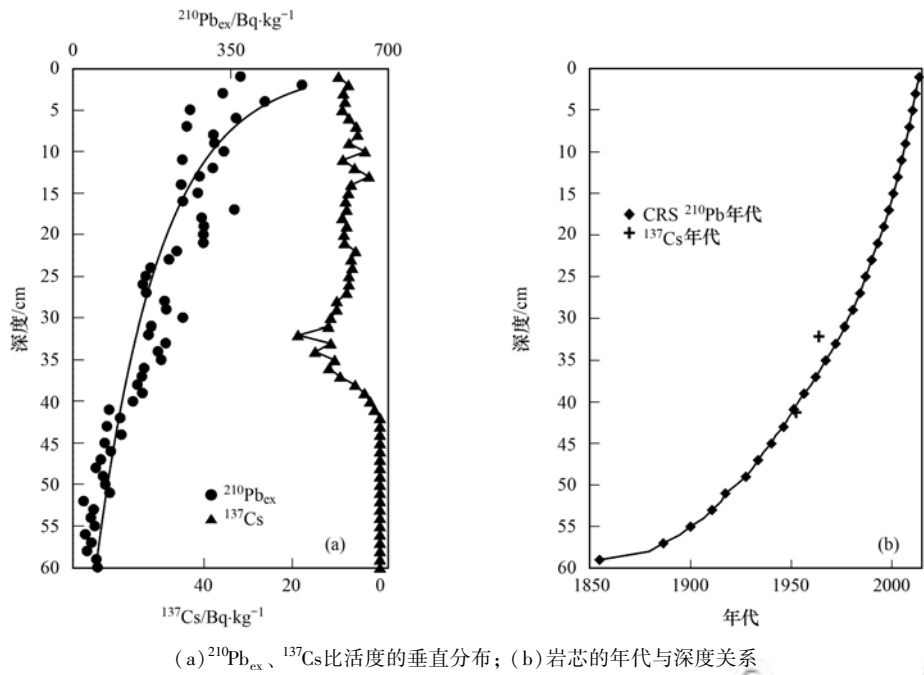
1.3 年代测定

²¹⁰Pb和¹³⁷Cs比活度采用美国EG & G Ortec公司生产的由高纯锗井型探测器(Ortec HPGe GWL)与Ortec 919型谱控制器计算机构成的16K多道分析器所组成的γ谱分析系统测定。该实验在中国科学院南京地理与湖泊研究所湖泊与环境国家重点实验室进行。

2 结果与讨论

2.1 网湖年代序列的建立

网湖沉积柱芯中过剩²¹⁰Pb(²¹⁰Pb_{ex})和¹³⁷Cs比活度随深度变化特征如图2所示。从图2(a)可以看出²¹⁰Pb_{ex}的比活度随深度呈指数递减,以²¹⁰Pb垂直剖面分布特征为依据,采用恒定放射性通量模式(CRS模式)计算沉积年代和速率,结果显示32 cm处约为1975年,41 cm处约为1951年,58 cm处年代结果为1980年,平均沉积速率为0.37 cm·a⁻¹。¹³⁷Cs蓄积峰(18.6 Bq·kg⁻¹)出现的深度为32 cm,对应1963年全球¹³⁷Cs散落高峰;深度41 cm处开始出现(1.2 Bq·kg⁻¹),对应1954年全球核实验开始^[28, 29]。CRS



(a) $^{210}\text{Pb}_{\text{ex}}$ 、 ^{137}Cs 比活度的垂直分布; (b) 岩芯的年代与深度关系

图2 网湖沉积岩芯中 $^{210}\text{Pb}_{\text{ex}}$ 、 ^{137}Cs 比活度的垂直分布及年代序列

Fig. 2 Distribution of $^{210}\text{Pb}_{\text{ex}}$, ^{137}Cs activities in Lake Wanghu sediment core and age model for the core

模式计算的网湖年代和 ^{137}Cs 定年有很好的-致性,定年结果如图2(b)所示。

2.2 网湖沉积物岩芯中正构烷烃的分布特征

正构烷烃广泛分布于细菌、藻类以及高等植物等生物体中,不同生物的正构烷烃具有不同的分布类型、碳数范围及主峰碳数。细菌和藻类等微生物正构烷烃碳数分布范围为 $n\text{-C}_{15} \sim n\text{-C}_{20}$,主峰碳为 $n\text{-C}_{15}$ 或 $n\text{-C}_{17}$,水生维管束植物的正构烷烃以 $n\text{-C}_{21}$ 、 $n\text{-C}_{23}$ 或 $n\text{-C}_{25}$ 为主峰,而大多数陆生维管

束植物表皮蜡质中含有很高比例的长链正构烷烃,以 $n\text{-C}_{27}$ 、 $n\text{-C}_{29}$ 或 $n\text{-C}_{31}$ 为主峰,具有显著的奇碳优势^[4, 5, 7, 8],因此,沉积物正构烷烃的碳数分布特征有明确的生物源指示意义。网湖沉积岩芯中正构烷烃碳数分布范围为 $n\text{-C}_{14} \sim n\text{-C}_{33}$ (图3),有单峰和双峰两种分布形式,单峰态的主峰碳为 $n\text{-C}_{17}$ 或 $n\text{-C}_{29}$,双峰态则有 $n\text{-C}_{17}$ 和 $n\text{-C}_{29}$ 两个峰值。高碳数正构烷烃($C_n \geq C_{22}$)相对丰度占总正构烷烃的70%以上,碳优势指数(carbon preferencee

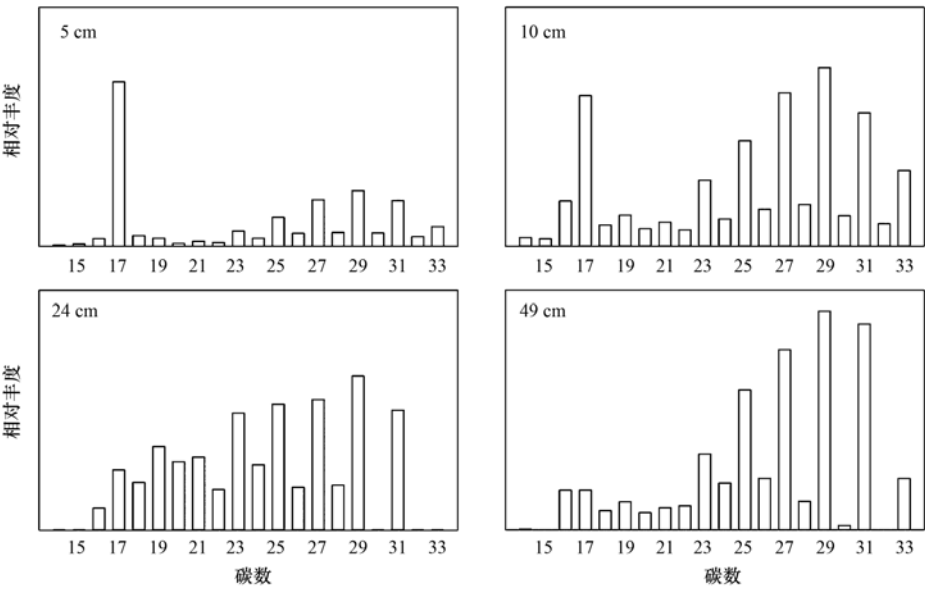


图3 网湖沉积岩芯中典型正构烷烃碳数分布

Fig. 3 Distributions of n -alkanes in typical samples of Lake Wanghu sediments

index, CPI) 为 1.9 ~ 14.6, 均值为 4.4, 具有明显的奇碳优势, 植物来源特征明显^[3], 而低碳数正构烷烃含量相对较低, 无明显的奇偶优势. 因此网湖沉积物长链正构烷烃主要来自维管束植物, 包括陆生以及水生维管束植物. 此外, 正构烷烃在垂向分布上表现出明显的阶段性变化特征: 底部以高碳数正构烷烃系列为主, 中部则逐渐过渡呈双峰分布模式, 高、低碳数系列含量均较高, 上部以低碳数同系物为主, 表明沉积物中有机质来源发生了明显的变化.

总正构烷烃的变化范围在 $0.1 \sim 1.8 \mu\text{g} \cdot \text{g}^{-1}$ 之

间, 平均值是 $0.4 \mu\text{g} \cdot \text{g}^{-1}$ (图 4). 20 世纪 50 年代以前, 总正构烷烃变化范围为 $0.1 \sim 0.2 \mu\text{g} \cdot \text{g}^{-1}$, 平均为 $0.2 \mu\text{g} \cdot \text{g}^{-1}$; 20 世纪 50 ~ 80 年代, 在 $0.1 \sim 0.2 \mu\text{g} \cdot \text{g}^{-1}$ 之间波动, 平均为 $0.1 \mu\text{g} \cdot \text{g}^{-1}$; 20 世纪 80 年代后在 $0.1 \sim 1.8 \mu\text{g} \cdot \text{g}^{-1}$ 之间波动, 平均为 $0.5 \mu\text{g} \cdot \text{g}^{-1}$. 总体上, 总正构烷烃在 20 世纪 80 年代以前, 含量较低, 之后含量明显增加, 并在表层达到最大值. 总有机质 (TOC) 的变化范围为 $0.4\% \sim 2.5\%$, 均值为 1.3% . 总正构烷烃的含量变化与 TOC 相似, 表现出显著相关 ($R^2 = 0.626$), 表明正构烷烃能较好地指示有机质变化.

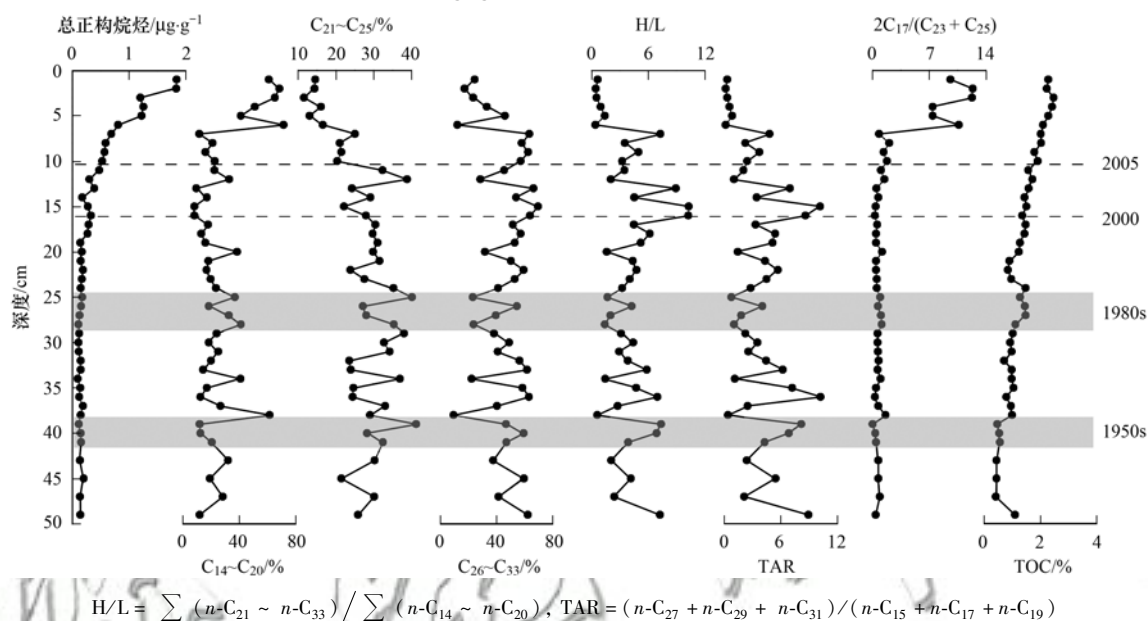


图 4 网湖沉积岩芯正构烷烃含量及其特征指数随深度的变化

Fig. 4 Depth profile of the *n*-alkanes content and derived proxies in Lake Wanghu sediment core

2.3 近百年来网湖沉积物中正构烷烃变化的环境意义

不同生物来源的正构烷烃组成不同, 因此不同链长正构烷烃的比例、H/L、TAR 可以用来反映来自于陆源高等植物和内源菌藻类正构烷烃的相对含量^[30~33], $2n-C_{17}/(n-C_{23} + n-C_{25})$ 比值可以揭示菌藻类和大型水生植物对沉积物有机质的相对贡献^[34]. 陆地木本植物来源的正构烷烃多以 $n-C_{27}$ 和 $n-C_{29}$ 为主, 草本植物以 $n-C_{31}$ 和 $n-C_{33}$ 为主, 因此, $2n-C_{31}/(n-C_{27} + n-C_{29})$ 比值可以用来表示沉积物中木本植物和草本植物的相对比例^[7, 8, 34]. 网湖沉积岩芯正构烷烃各特征参数随深度变化如图 4 所示, 短链 ($n-C_{14} \sim n-C_{20}$)、中链 ($n-C_{21} \sim n-C_{25}$) 和长链 ($n-C_{26} \sim n-C_{33}$) 正构烷烃分别占总含量的 $8.3\% \sim 71.2\%$ (平均值为 27.0%)、 $11.5\% \sim 41.2\%$ (平均值为 27.3%) 和 $9.7\% \sim 69.5\%$ (平均值为 45.7%), H/L

的变化范围在 $0.4 \sim 10.3$ 之间, 平均值是 3.9 , TAR 的变化范围在 $0.2 \sim 10.2$ 之间, 平均值是 3.7 , $2n-C_{17}/(n-C_{23} + n-C_{25})$ 比值变化范围为 $0 \sim 12.4$, 平均值是 2.0 , 表明总体上浮游藻类对沉积物中有机质的贡献较少, 这与网湖是草型湖的特征相符; $2n-C_{31}/(n-C_{27} + n-C_{29})$ 比值为 $0.2 \sim 1.7$, 平均值为 1.0 , 表明网湖沉积物的外源有机质中木本植物占优势, 与孢粉鉴定结果一致^[26].

根据网湖沉积物中正构烷烃参数变化, 将近百年来网湖环境变化大致分为 3 个阶段如下所示.

20 世纪 50 年代以前, 总正构烷烃、TOC 含量稳定, 表明湖泊初级生产力较低且稳定. H/L、TAR 值较大, $2n-C_{17}/(n-C_{23} + n-C_{25})$ 比值平均值是 0.7 , 表明沉积物中有机质以大型水生植物和陆生高等植物为主, 而浮游藻类含量相对较少, 反映了湖泊的营养程度低, 水体质量相对较好. 当时网湖为通江湖泊,

受长江水位影响大,多次发生的大洪水,造成江水携带而至的外源有机质增多,而湖泊水体频繁的交换,利于水生植物的生长。

20 世纪 50~80 年代的 TOC 值较前一阶段略有上升,反映了湖泊环境相较前一阶段有所变化。长链正构烷烃的比例、H/L 和 TAR 值在 20 世纪 60 年代出现高值后明显下降,短链和中链正构烷烃比例的变化趋势则相反, $2n-C_{17}/(n-C_{23} + n-C_{25})$ 比值的变化范围为 0~1.6,平均值是 0.6,比值仍处于较低范围内,表明外源有机质减少,大型水生植物和藻类有机质增加,浮游藻类如淡水盘星藻和双星藻在这个阶段开始频繁出现^[26],但浮游藻类对有机质的贡献相较于大型水生植物较少。20 世纪 60 年代长链正构烷烃的比例、H/L 和 TAR 的高值与当时阳新县较多的洪水、大规模的水利建设,围湖造田,导致大量陆源碎屑物入湖有关^[24, 35];之后的下降与江湖关系的变化有关,富池大坝建成后阻断了网湖与长江的天然联系,使得网湖由原来的通江湖泊演变为水位由人工控制的季节性封闭湖泊,江水携带的外源有机质减少。

20 世纪 80 年代以来,尤其是 2000 年后总正构烷烃明显升高,在表层达到最高,表明正构烷烃输入明显增加,结合较高的 TOC 含量,可见此沉积时段湖泊的营养水平明显升高。中链和长链正构烷烃的比例、H/L 和 TAR 值先升高再降低,短链正构烷烃的比例、 $2n-C_{17}/(n-C_{23} + n-C_{25})$ 比值和总正构烷烃的变化趋势一致,表明大型水生植物和陆生高等植物对沉积物中有机质的贡献先增大后减少,而藻类有机质的含量明显增加,反映了湖泊环境发生明显变化。20 世纪 80、90 年代长江流域洪水频发,外源有机质输入增加,且网湖变为封闭型湖泊后水体滞留时间延长,再加上随着长江流域经济的快速发展,人类活动强度的明显增强,对网湖周边生态环境影响变大,外源营养盐及陆源有机物的输入增加,导致湖泊水体的营养水平明显升高,浮游藻类生长加快。湖北省 2014 年环境调查显示网湖的营养水平达到中富营养^[36]。

3 结论

网湖沉积岩芯中的正构烷烃研究表明,近百年来沉积物中正构烷烃主要来源于陆源高等植物和大型水生植物,而陆源有机质输入以木本植物为主,但近年来湖泊浮游藻类贡献有明显增加趋势。网湖沉积物中正构烷烃表现为高碳数占优势, $2n-C_{31}/$

$(n-C_{27} + n-C_{29})$ 比值整体较低, $2n-C_{17}/(n-C_{23} + n-C_{25})$ 比值在 2000 年后呈现快速升高趋势。根据正构烷烃参数将网湖过去 100 年来的环境变化划分为 3 个时期。20 世纪 50 年代以前,湖泊受长江水位变化影响较大,湖泊水体交换频繁,水体营养水平低,外源陆生高等植物和大型水生植物是湖泊主要的有机质来源,体现在沉积物总正构烷烃、TOC 和 $2n-C_{17}/(n-C_{23} + n-C_{25})$ 比值较低且稳定,沉积物长链正构烷烃的比例、H/L 和 TAR 较高。20 世纪 50~80 年代,湖泊受长江水位影响较弱,湖泊沉积物有机质中外源陆生植物贡献下降,湖泊藻类和大型水生植物贡献增加,湖泊营养水平升高,表现在沉积物长链正构烷烃的比例、H/L 和 TAR 值下降,中、短链正构烷烃的比例升高,TOC 值增加。20 世纪 80 年代以来,人类活动增强,陆源有机物输入增加,引起湖泊营养水平明显升高,导致湖泊内源浮游藻类生长加快,体现在这一阶段中、长链正构烷烃的比例、H/L 和 TAR 值先升高后下降,总正构烷烃、低链正构烷烃的比例和 $2n-C_{17}/(n-C_{23} + n-C_{25})$ 值明显升高,尤其是 2000 年后变化显著。

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《环境科学》多项引证指标名列前茅

2016 年 10 月 12 日,中国科学技术信息研究所在中国科技论文统计结果发布会上公布了 2015 年度中国科技论文统计结果. 统计结果显示《环境科学》2015 年度总被引频次 8 844,影响因子 1.617,多项引证指标位居环境科学技术及资源科学技术类科技期刊前列.



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