北京城区PM_{2.5}各组分污染特征及来源分析

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洪泽湖溶解态有机质与重金属汞的结合特性

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摘要: 水体中溶解态有机质(DOM)能够与重金属络合形成复合化合物,进而影响水环境中重金属的迁移转化、生物可利用性和毒性. 通过荧光光谱结合不同的光谱分析方法研究了洪泽湖不同季节水体 DOM 与重金属汞(Hg)的结合特性. 结果表明,春季 DOM 以类蛋白质为主,秋季则以类腐殖质为主,洪泽湖秋、冬季蓄水导致 DOM 性质存在显著差异. 同步荧光光谱结合二维相关光谱分析结果表明,洪泽湖春季 DOM 与Hg(II)的结合强度依次为:360~425 nm > 290 nm > 300~350 nm,秋季 DOM 与Hg(II)的结合强度依次为:290 nm > 315~355 nm > 365~380 nm > 380~465 nm. 三维荧光光谱和荧光淬灭实验表明,洪泽湖水体 DOM 的5种荧光组分均与Hg(II)有不同程度地络合. Ryan-Weber 模型拟合结果进一步表明,春季 DOM 类腐殖质组分与Hg(II)结合能力强于类蛋白质,秋季则是类蛋白质组分的结合能力强于类腐殖质. 总体而言,洪泽湖秋、冬季蓄水能够显著改变 DOM 的性质,从而影响 DOM 与重金属Hg(II)的结合特性,可能会增强洪泽湖水体中 Hg 的生物可利用性和毒性.

关键词: 溶解态有机质(DOM); 汞(Hg); 荧光光谱; 荧光淬灭; 洪泽湖

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Binding Affinity Between Heavy Metal Hg and Dissolved Organic Matter in Hongze Lake

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Abstract: Dissolved organic matter (DOM) can form complex compounds with heavy metals and then affect the transport, transformation, bioavailability, and toxicity of heavy metals in aquatic environments. Here, fluorescence spectroscopy combined with various spectral analysis methods were conducted to study the binding affinity between heavy metal Hg and seasonal dissolved organic matter in Hongze Lake. DOM was dominated by protein-like substances in spring, whereas DOM in autumn was dominated by humic-like substances. These results indicated that lake impoundment during autumn and winter resulted in significant variations in DOM compositions in Hongze Lake. Two-dimensional correlation spectroscopy analysis of synchronous fluorescence spectra revealed that the binding sequence to Hg(II) was 360-425 nm > 290 nm > 300-350 nm for spring DOM and 290 nm > 315-355 nm > 365-380 nm > 380-465 nm for autumn DOM. Fluorescence quenching characteristics showed a relatively different trend for five fluorescent components. The results of the Ryan-Weber model further indicated that the binding affinity with Hg(II) for spring humic-like components was higher than that for protein-like components. However, the opposite binding affinity between humic- and protein-like components and Hg(II) was observed for autumn DOM. In general, impoundment could significantly alter the DOM composition and the binding affinity between DOM and heavy metal Hg and may eventually increase the bioavailability and toxicity of Hg in Hongze Lake.

Key words: dissolved organic matter (DOM); mercury (Hg); fluorescence spectroscopy; fluorescence quenching; Hongze Lake

溶解态有机质(dissolved organic matter, DOM)是水环境中普遍存在且非常重要的物质之一,其含有丰富的碳、氮和磷等生源要素,不仅是全球碳循环的重要组成部分之一,也是生态系统中能量和物质循环的重要桥梁^[1,2]. 此外,DOM作为重金属等污染物的载体,能够通过微生物作用、光化学反应、矿化和结合等一系列环境行为,影响污染物的溶解性、生物可利用性、迁移和生物毒性等生物地球化学过程^[3-5]. 因此,DOM具有重要的生态和环境意义,其丰富的环境行为和生态环境效应是水环境领域和生物地球化学的研究热点.

DOM具有丰富的官能团,如羧基和酚羟基等,能够以静电结合和有机络合等方式与水体中的金属离子形成复合化合物,进而影响重金属的毒性和生物有效性等^[6-8]. 国内外学者针对水环境中DOM与

重金属的结合开展了一系列的研究^[9-11]. Xu等^[9]的研究结果表明,草型湖泊沉积物有机质对重金属有更多的结合点位和更强的结合能力,相比于藻型湖泊沉积物,能够减弱重金属的生物毒性. Chen等^[10]的研究发现,富里酸和亲水性有机酸能够通过络合促进藻类对铜的吸附,进而影响其毒性和生物有效性. 由于DOM与重金属离子发生作用后,其结构和光学特征也会发生相应地变化,因此光谱分析方法被广泛地应用,以深入了解DOM与重金属的相互作用机制^[12-14].

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汞(Hg)是一种有毒重金属元素,水环境中的Hg可以通过微生物和光化学等过程转化成甲基汞,对人类和水生生物的健康造成严重的危害^[15,16]. DOM的多种活性官能团可以与Hg离子结合,影响水环境中Hg的赋存形态,进而影响汞的形态转化^[17,18]. 此外DOM也可通过微生物群落和羟基自由基等间接影响汞的形态转化^[19~21],因此开展DOM与Hg的相互作用有助于丰富水环境中Hg的生物地球化学过程. 本文以大型浅水湖泊洪泽湖DOM为研究对象,利用同步荧光光谱法和三维荧光光谱法,结合二维相关光谱分析,分析不同季节洪泽湖DOM不同组分与Hg的结合特性,以期为进一步了解DOM在湖泊重金属毒性和生物有效性方面所起的作用提供一定的科学依据和研究基础.

1 材料与方法

1.1 研究区域概况和样品采集

洪泽湖(33°06' ~ 33°40' N, 118°10' ~ 118°52' E),不仅是我国第四大淡水湖,也是淮河流域最大的湖泊,兼具蓄洪、灌溉、航运、水产养殖、饮用和生态保护等多项功能,同时也是南水北调东线工程

重要的调蓄性湖泊,具有明显的水库特征^[22,23]. 洪泽湖入湖河流集中在西部,年平均入湖流量330亿m³,其中淮河为最大入湖河流,入湖水量占总入湖量的70%以上. 出湖河流分布在东部,其中入江水道为淮河和洪泽湖的主要泄洪道,60%~70%的湖水经由入江水道流入长江. 洪泽湖属于过水型湖泊,年均换水率达11次之多. 为了更好地调节利用水资源,洪泽湖3条主要出湖河流上建设了人工闸门,每年5~10月开启闸门以满足防洪排涝和农业灌溉等用水需求,而每年11月~次年4月则关闭闸门蓄水以满足调水、饮水和航运等需求^[11,24].

本研究根据洪泽湖湖盆形态、水文条件和出入湖湖口位置,选择淮河入湖区、西部湖区、南部湖湾区和湖心区4个采样点(表1),分别在2016年4月和11月使用Rutter分层水样采集器采集表层水,采集的水样使用0.22 μm聚醚砜树脂滤膜过滤,并置于棕色聚乙烯瓶中冷藏避光保存待测. 使用美国HACH多参数水质检测仪现场测定水样的pH和溶解氧(dissolved oxygen, DO),使用德国Elementar Liqui-TOC总有机碳分析仪测定溶解态有机碳(dissolved organic carbon, DOC)含量.

表1 洪泽湖采样点坐标和水样理化性质

Table 1 Location of Hongze Lake sampling sites and physical-chemical properties of samples

点位	点位经纬度		pH		DO/mg·L ⁻¹		ρ(DOC)/mg·L ⁻¹	
	经度(E)/(°)	纬度(N)/(°)	4月	11月	4月	11月	4月	11月
HZ1	118.45499	33.21663	8.55	8.32	9.92	9.46	14.60	25.93
HZ2	118.74515	33.27332	8.50	8.13	10.01	9.14	13.61	16.27
HZ3	118.72760	33.15653	8.70	8.03	9.75	8.32	13.81	16.43
HZ4	118.61655	33.22010	8.62	7.99	9.64	8.07	13.53	16.38

1.2 实验方法与指标测定

1.2.1 重金属结合实验

将不同体积的浓度为0.025 mol·L⁻¹的Hg(NO₃)₂储备液滴定至滤后水样中,使得水样中Hg(II)最终浓度分别为0、5、10、15、20、30、40、50、75、100、150、200和250 μmol·L⁻¹. 为避免浓度影响每次滴加的量均少于水样的5%^[9]. 在室温条件下恒温避光振荡反应24 h,实验后立即测定样品的荧光光谱.

1.2.2 荧光光谱的测定与分析

采用日本Hitachi公司的F-7000荧光光度计测定Hg(II)添加前后水样的三维荧光光谱和同步荧光光谱,光源为150 W连续输出氙弧灯. 其中三维荧光光谱测定激发波长范围为230~450 nm,步长5 nm. 发射波长范围为325~550 nm,步长1 nm. 激发和发射波长的夹缝宽均为5 nm,扫描速度为12 000 nm·min⁻¹. 使用Milli-Q超纯水作为空

白样品用于消除水拉曼效应造成的荧光光谱拉曼峰. 使用MATLAB软件利用平行因子分析法对三维荧光光谱进行解析^[25]. 本文采用硫酸奎宁单位(QSE)校准并归一所有三维荧光光谱的荧光强度^[15].

同步荧光光谱的测定激发波长为250~500 nm,设定发射波长与激发波长的恒定间隔为60 nm. 将重金属浓度作为外扰因素,利用“2D shige”软件建立同步和异步二维荧光相关光谱^[26].

1.2.3 荧光淬灭曲线拟合模型

为了综合评估DOM不同组分与重金属的结合作用情况,使用Ryan-Weber模型对荧光淬灭曲线进行非线性拟合^[27]. 该模型假设结合点位与重金属离子按照1:1的形式结合成有机金属复合物,从而引起荧光淬灭反应^[28]. 拟合方程如下:

$$F = F_0 + (F_{ML} - F_0) \left(\frac{1}{2K_M C_L} \right) \times$$

$$\left[1 + K_M C_L + K_M C_M - \sqrt{(1 + K_M C_L + K_M C_M)^2 - 4K_M^2 C_M C_L} \right]$$

式中, F 为不同重金属浓度 C_M 下的荧光峰强, F_0 为没有加入重金属的初始荧光峰强, F_{ML} 为重金属浓度增大到一定值, 荧光峰强不再发生变化的荧光峰限值, K_M 和 C_L 分别为条件稳定常数和最大结合容量. 条件稳定常数 ($\lg K_M$) 用来表征体系中 DOM 与重金属结合潜能的稳定性. 用 C_L 除以 DOC 浓度得到的值 ($\text{mmol} \cdot \text{g}^{-1}$) 即为可表征 DOM 与重金属的结合容量 (CC_c).

2 结果与讨论

2.1 溶解态有机质同步荧光光谱随 $\text{Hg}(\text{II})$ 增加的变化

洪泽湖 DOM 的同步荧光光谱荧光强度随 $\text{Hg}(\text{II})$ 浓度增加的变化情况如图 1 所示. 由图 1 (a) 和 1 (b) 可以看出, 春季 (4 月) 洪泽湖水体中 DOM 分别在 280 nm 和 350 nm 处存在显著的特征峰, 秋季 (11 月) 水体 DOM 则在 350 nm 存在一个显著的特征峰. 有研究表明, 波长在 200 ~ 300 nm 和

(a) 同步荧光光谱-4月, (b) 同步荧光光谱-11月, (c) 同步二维相关光谱-4月, (d) 同步二维相关光谱-11月, (e) 异步二维相关光谱-4月, (f) 异步二维相关光谱-11月

图 1 洪泽湖溶解态有机质同步荧光光谱随 $\text{Hg}(\text{II})$ 浓度变化情况和其同步和异步二维相关光谱图

Fig. 1 Changes in the SF spectra of dissolved organic matter from Hongze Lake upon addition of $\text{Hg}(\text{II})$ and its synchronous and asynchronous 2D-COS maps

300 nm 以上的特征峰分别归因于类蛋白质和类腐殖质^[13,28]. 这表明洪泽湖春季水体 DOM 以类蛋白质和类腐殖质为主, 而秋季水体 DOM 则以类腐殖质为主, 与已有的研究结果相同^[11,29]. 此外, 秋季水体 DOM 荧光强度和 DOC 含量均远高于春季水体(表 1 和图 1), 这主要是由于夏、秋季上游淮河陆源性 DOM 大量输入造成的^[11].

随着 $c[\text{Hg}(\text{II})]$ 从 $0 \mu\text{mol}\cdot\text{L}^{-1}$ 增加至 $250 \mu\text{mol}\cdot\text{L}^{-1}$, DOM 不同波长的荧光强度均呈现明显的下降趋势[图 1(a) 和 1(b)]. 这一结果表明 $\text{Hg}(\text{II})$ 与 DOM 中不同组分都发生了结合作用. 为了进一步掌握 DOM 与重金属 Hg 的结合特性, 利用二维相关光谱解析同步荧光光谱以确定 $\text{Hg}(\text{II})$ 离子与 DOM 的结合点位和结合顺序, 结果如图 1(c) ~ 1(f) 所示.

如图 1(c) 和 1(d) 所示, 春、秋季洪泽湖 DOM 同步二维相关光谱图均在 350 nm/350 nm 和 290 nm/290 nm 处存在正值的自峰, 在 290 nm/350 nm 存在一个交叉峰. 此外, 秋季水体 DOM 的同步二维相关光谱的自峰峰强远高于春季水体, 表明, 洪泽湖秋季水体 DOM 在滴加 $\text{Hg}(\text{II})$ 后荧光光强更容易降

低, 荧光淬灭效应更加显著. 如图 1(e) 和 1(f) 所示, 异步二维光谱图的峰值呈对角线分布, 在对角线下方均存在正值峰和负值峰. 根据 Noda 法则^[26], 洪泽湖春季 DOM 与 $\text{Hg}(\text{II})$ 的结合强度为: $360 \sim 425 \text{ nm} > 290 \text{ nm} > 300 \sim 350 \text{ nm}$, 秋季 DOM 与 $\text{Hg}(\text{II})$ 的结合强度为: $290 \text{ nm} > 315 \sim 355 \text{ nm} > 365 \sim 380 \text{ nm} > 380 \sim 465 \text{ nm}$. 异步二维相关光谱的结果表明, 洪泽湖春季 DOM 类腐殖质对 $\text{Hg}(\text{II})$ 的亲合力强于类蛋白质, 而秋季 DOM 则是类蛋白质的亲合力强于类腐殖质.

2.2 洪泽湖溶解态有机质三维荧光光谱解析

随着 $c[\text{Hg}(\text{II})]$ 浓度由 $0 \mu\text{mol}\cdot\text{L}^{-1}$ 逐渐增加至 $250 \mu\text{mol}\cdot\text{L}^{-1}$, 春、秋季水体 DOM 不同波长荧光团的荧光峰值均发生了明显地下降(图 2), 说明在 $\text{Hg}(\text{II})$ 的作用下 DOM 发生了淬灭效应, 这一结果与图 1 同步荧光光谱结果相同. 为了进一步了解 DOM 中不同组分与 $\text{Hg}(\text{II})$ 的结合特性, 利用平行因子方法对三维荧光光谱进行解析, 基于残差分析和分半验证结果共解析出 5 个荧光组分. 通过对这 5 个荧光组分的光谱参数和 OpenFluor 在线光谱数据库中公开的 PARAFAC 模型进行比较, 发现 Tucker 一致性系数均超过 0.95, 认定此次解析结果

(a) 4 月, $c[\text{Hg}(\text{II})] = 0 \mu\text{mol}\cdot\text{L}^{-1}$, (b) 11 月, $c[\text{Hg}(\text{II})] = 0 \mu\text{mol}\cdot\text{L}^{-1}$, (c) 4 月, $c[\text{Hg}(\text{II})] = 250 \mu\text{mol}\cdot\text{L}^{-1}$, (d) 11 月, $c[\text{Hg}(\text{II})] = 250 \mu\text{mol}\cdot\text{L}^{-1}$

图 2 洪泽湖溶解态有机质与 $\text{Hg}(\text{II})$ 结合三维荧光光谱图

Fig. 2 Changes in excitation-emission matrix spectroscopy of dissolved organic matter from Hongze Lake binding with $\text{Hg}(\text{II})$

是可信的^[30,31].

根据光谱特征将 5 个荧光组分分成两大类,其中 3 个成分(C1、C2 和 C3)可归一为类腐殖质荧光成分,而其余 2 个成分(C4 和 C5)为类蛋白质荧光成分. 其中,C1 [$E_x/E_m = (230(340) \text{ nm}/425 \text{ nm})$] 和 C3 [$E_x/E_m = 260(280) \text{ nm}/460 \sim 500 \text{ nm}$] 被普遍认为是陆源类腐殖质成分,具有高芳香性和高分子质量的特点^[32~34]. C2 [$E_x/E_m = 245(305) \text{ nm}/405 \text{ nm}$] 的最大激发和发射波长类似于微生物作用产生的海源腐殖质 M 峰^[33~35]. C4 [$E_x/E_m = (230(290) \text{ nm}/350 \text{ nm})$] 和 C5 [$E_x/E_m = 275((230) \text{ nm}/320 \text{ nm})$] 都是典型的类蛋白质荧光成分,其中 C4 为色氨酸,C5 为酪氨酸^[36~38].

2.3 洪泽湖溶解态有机质不同组分与 Hg(II) 的结合特性

为了对比研究 DOM 各组分在不同 Hg(II) 浓度

下的淬灭程度,图 3 分别作出各荧光组分在不同 Hg(II) 浓度作用下的荧光淬灭曲线. 从中可见,组分 C1 和 C4 在避光条件振荡 24 h 后呈现明显的淬灭效应,最大荧光强度随着 Hg(II) 浓度的升高而降低,但 C2、C3 和 C5 这 3 种组分受 Hg(II) 的淬灭影响很小,甚至出现荧光强度整体增强的变化趋势. 荧光组分强度的增强可能是由于重金属 Hg(II) 与有机质结合,改变了有机质的结构造成的,这一现象与其他研究相类似^[19,39]. 显著差异的淬灭曲线表明,有机质不同组分与重金属 Hg(II) 的结合特性大不相同,这一结果与同步荧光光谱结果相类似.

利用 Ryan-Weber 模型对 Hg(II) 作用下有机质不同组分的荧光淬灭曲线进行非线性拟合. 组分 C2、C3 和 C5 虽然整体呈现荧光强度下降的趋势,但 3 个组分在不同浓度下的荧光强度均波动较大,模型的拟合结果较差. 因此,本文对 DOM 组分 C1

图 3 洪泽湖 DOM 不同组分与 Hg(II) 结合荧光淬灭曲线

Fig. 3 Fluorescence quenching curve between Hg(II) and different fluorescence components from Hongze Lake DOM

和 C4 淬灭曲线进行非线性模型拟合, 结果如表 2 所示。

表 2 通过 Ryan-Weber 模型确定的荧光组分与 Hg(II) 的结合参数 ($n=13$)

Table 2 Values of binding parameters for fluorescence components with Hg(II) determined by Ryan-Weber model ($n=13$)

荧光组分	采样点	春季(4月)			秋季(11月)		
		$\lg K_M$	CC_c	R^2	$\lg K_M$	CC_c	R^2
C1	HZ1	4.41	2.94	0.97	4.33	1.96	0.95
	HZ2	4.60	1.84	0.95	4.51	1.84	0.91
	HZ3	4.54	2.21	0.95	4.72	1.18	0.95
	HZ4	4.63	2.26	0.96	3.80	14.20	0.97
C4	HZ1	4.32	3.63	0.93	3.98	4.94	0.98
	HZ2	4.47	2.56	0.97	5.10	0.45	0.77
	HZ3	4.48	2.37	0.97	5.02	0.57	0.94
	HZ4	4.58	1.52	0.88	5.10	0.57	0.84

由表 2 可以看出, 春秋两季洪泽湖 DOM 与 Hg(II) 的结合稳定常数 $\lg K_M$ 范围为 3.80 ~ 5.10, 与其他研究结果相符^[40,41]。春季水体 DOM 荧光组分 C1 的 $\lg K_M$ 值均要高于荧光组分 C4, 表明春季 DOM 中类腐殖质与重金属 Hg(II) 的亲合力要强于类蛋白质。秋季水体中 DOM 荧光组分 C1 的 $\lg K_M$ 值则低于荧光组分 C4 (HZ1 除外), 表明秋季 DOM 中类蛋白质与重金属 Hg(II) 的亲合力要强于类蛋白质。不同季节 DOM 荧光组分与 Hg(II) 结合稳定常数 $\lg K_M$ 结果与异步二维相关光谱结合顺序结果相符(图 1)。本实验结果进一步表明, 二维相关光谱结合荧光光谱能够很好地应用于 DOM 与重金属 Hg(II) 的结合研究中。

DOM 的性质变化对其与重金属的结合有着显著的影响^[42~44]。例如, Li 等^[42]的研究发现太湖藻类暴发期间富里酸与 Cu(II) 的结合能力要远高于非暴发期。有研究表明, 春季洪泽湖陆源 DOM 腐质类组分 C1 在秋季蓄水期间通过微生物和光化学等作用被转化为自生 DOM 蛋白类 C4^[11]。本文结果中春季 DOM 组分 C1 与 Hg(II) 的结合能力($\lg K_M$ 值)大部分高于秋季水体 DOM, 而类蛋白质(C4)的结合能力则大部分低于秋季水体(表 2)。与此同时, 春季水体中 DOM 组分 C1 和 C4 的结合容量要高于秋季水体。春、秋季 DOM 与重金属 Hg(II) 显著差异的结合特性表明, 洪泽湖秋冬季蓄水期间 DOM 性质的变化可能会影响水体中重金属 Hg 的形态变化, 进而影响 Hg 的生物可利用性和毒性。此外, Lescord 等^[45]的研究表明, 生物中甲基汞的含量与内源性有机质含量呈显著正相关, Jiang 等^[17]的研究认为内源性有机质能够影响微生物群落活性进而刺激环境中甲基汞的形成。洪泽湖蓄水期间 DOM 性质的变化可能会增强水体中 Hg 的生物可利用性和毒性, 因此, 蓄水期间应当加强水环境中重金属 Hg 的监测以保障用水安全。

3 结论

(1) 春季洪泽湖 DOM 以类蛋白质和类腐殖质物质为主, 秋季洪泽湖 DOM 则以类腐殖质物质为主。

(2) 洪泽湖春季 DOM 与 Hg(II) 的结合强度依次为: 360 ~ 425 nm > 290 nm > 300 ~ 350 nm, 秋季 DOM 与 Hg(II) 的结合强度依次为: 290 nm > 315 ~ 355 nm > 365 ~ 380 nm > 380 ~ 465 nm。

(3) 洪泽湖水体 DOM 包括 3 种类腐殖质和 2 种类蛋白质荧光组分。其中春季 DOM 类腐殖质组分 C1 与 Hg(II) 结合能力强于类蛋白质 C4, 秋季则是类蛋白质组分 C4 强于类腐殖质 C1。

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