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分层型水源水库溶解性有机物性质及其膜污染特性

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摘要: 利用激发-发射矩阵(EEM)荧光光谱和紫外吸收光谱研究了深水型水源水库热分层期溶解性有机物(DOM)性质及其膜污染特性随水深的变化。结果表明, 水体热分层导致 DOM 质量浓度和性质也表现出分层特征。变温层受光化学降解影响较大, DOM 质量浓度较低, 同时受藻类等浮游植物分泌的内源有机物影响, DOM 芳香度较低, 类富里酸有机物(C1 组分)和类腐殖酸有机物(C2 组分)荧光强度较低, 但类色氨酸有机物(C3 组分)荧光强度较高; 斜温层 DOM 受径流输入影响较大, DOM 质量浓度和芳香度较高, C1 和 C2 组分荧光强度较高。膜污染方面, 变温层 DOM 造成的总污染最大, 但可逆性较好, 斜温层和等温层 DOM 造成的总污染较低, 但可逆性较差; 对超滤过程中不同荧光组分迁移的分析表明, 超滤膜对 C3 组分截留率较高, 但反冲洗对被截留的 C3 组分去除效果较好, 而被膜截留的 C1 和 C2 组分较难被反冲洗去除。

关键词: 分层型水库; 溶解性有机物(DOM); 紫外吸收光谱; 激发-发射矩阵(EEM)荧光光谱; 膜污染

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Characteristics and Fouling Potential of Dissolved Organic Matter in a Stratified Source Water Reservoir

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Abstract: Quantity and quality of dissolved organic matter (DOM) in source water are critical factors affecting both the drinking water quality and operation of the water treatment process. As major water sources for many cities, deep reservoirs usually suffer seasonal thermal stratification, which has a significant influence on the fate and transport of many contaminants including iron, manganese, ammonia and DOM. This study focuses on the variation of properties and the fouling potential of DOM in a deep-source water reservoir during the thermal stratification period. Excitation-emission matrix (EEM) fluorescence spectra and ultraviolet absorption spectra were used to characterize the DOM. The results indicate that the quantity and quality of DOM vary with the water depth due to thermal stratification. The DOC of epilimnion is lower than that of the metalimnion and hypolimnion due to photochemical degradation. Moreover, organic matter released by phytoplankton, such as algae, play an important role in the DOM composition. Therefore, the DOM of the epilimnion exhibits a lower aromaticity, lower fluorescence intensities of Component 1 (i. e., fulvic-like substance) and Component 2 (i. e., humic-like substance), and higher fluorescence intensity of Component 3 (i. e., tryptophan-like substance). The DOM of the metalimnion is dominated by runoff input and therefore the concentration, aromaticity, and fluorescence intensities of Components 1 and 2 are higher. In terms of membrane fouling, total fouling caused by the DOM of the epilimnion is the largest, but its reversibility is better. The DOM of the metalimnion and hypolimnion results in lower total fouling but poor reversibility. The analysis of the fate of different fluorescence components during ultrafiltration suggests that the UF shows a relative high rejection rate for Component 3, which could be readily removed by backwash, whereas Component 1 and Component 2 retained by the membrane are difficult to be removed by backwash.

Key words: stratified reservoir; dissolved organic matter (DOM); UV absorbance; excitation-emission matrix (EEM) fluorescence spectra; membrane fouling

水源水中溶解性有机物(dissolved organic matter, DOM)是影响饮用水水质安全和水处理工艺运行的重要因素。DOM 不但是水中色度和臭味的重要来源, 还是氯化消毒副产物的主要前体物, 对饮用水的感官性状和化学安全性都有着重要影响^[1]。此外, 在饮用水输配过程中部分 DOM 能够作为微生物生长的基质, 影响着饮用水的生物安全性^[2]。从饮用水处理工艺运行的角度来看, DOM 一方面是混凝、吸附、膜过滤等工艺单元的去

除, 另一方面 DOM 会影响混凝剂投量, 与微量污染物竞争吸附位, 导致膜污染^[3, 4]。

水库作为许多城市的主要供水水源, 其水力特征与河流有明显差异。对于深水型水库, 太阳辐射

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和热量传递的差异会导致水温和密度随水深而变化,从而形成水体热分层,从上至下分别是变温层、斜温层和等温层^[5].热分层期水体密度差阻碍着上下水体间物质的交换,底部水体无法得到大气复氧,在还原性物质和微生物等的耗氧作用下溶解氧会逐渐降低,进入缺氧甚至厌氧状态,引起水-沉积物界面处氮、磷、铁、锰、有机物等的形态转化;与此同时,在适宜的光照、温度、营养盐等条件下表层水体会出现藻类等浮游植物的大量繁殖,并存在一定的光化学降解作用;此外,入库径流的外源输入对水质也有重要影响^[6~8].目前,关于深水型水库 DOM 的来源、特征及其迁移转化已有一定的研究,例如,Chen 等^[9]对近年来关于沉积物中 DOM 的研究文献进行了全面综述,分析了沉积物与上覆水 DOM 在质量浓度、性质等方面的异同;Zhou 等^[1]考察了入库流量变化对大型水源水库中发色 DOM 组成和性质的影响;黄廷林等^[10]利用紫外光谱和三维激发-发射矩阵(excitation-emission matrix, EEM)荧光光谱分析了冬季混合期两个深水型水库 DOM 的性质,说明了 DOM 来源和特征与流域土地利用的关系;黄廷林等^[11]和方开凯等^[12]通过光谱分析研究了热分层期库区内不同位置表层 DOM 的来源和特征.但上述研究主要关注 DOM 在流域内以及水-沉积物界面的迁移转化过程,关于 DOM 特性对水体热分层的响应特征及其影响鲜有报道.

黑河金盆水库总库容 2.0 亿 m^3 ,有效库容 1.77 亿 m^3 ,是西安市主要饮用水水源,该水库为峡谷型深水水库,主库区平均水深 60~95m,每年 7~9 月会形成稳定的热分层^[13].本研究在监测热分层期水温和溶解氧随水深变化的基础上,利用紫外吸收光谱和 EEM 考察变温层、斜温层和等温层 DOM 特性,通过平板膜短期实验考察 DOM 膜污染特征,以期为分层型水源水库和相应自来水管的运行调度提供依据.

1 材料与方法

1.1 样品的采集与预处理

2016 年 7~9 月在黑河金盆水库引水塔附近使用深水采样器采集变温层(水深 0.5 m)、斜温层(水深 20 m)和等温层(水深 60 m)水样,水样采集后 12 h 内带回实验室用 0.45 μm 玻璃纤维滤膜过滤,过滤后水样保存在 4 $^{\circ}\text{C}$ 的冰箱内,3 d 内完成所有实验.

1.2 实验仪器与方法

使用 TOC-L-CPN 总有机碳分析仪(日本,岛

津)测定溶解性有机碳(DOC).使用 F-3900 紫外分光光度计(日本,日立)和 1 cm 石英比色皿测定紫外吸光度.采用 F-7000 荧光光谱仪(日本,日立)测定 EEM 光谱,激发波长范围为 200~400 nm,发射波长范围为 250~500 nm,激发光和发射光狭缝宽度均为 5 nm,扫描速度为 12 000 $\text{nm}\cdot\text{min}^{-1}$.

DOM 是由结构和性质差异很大的许多物质构成的混合物,因此测得的 EEM 光谱实际上是不同荧光组分相互叠加的结果.平行因子分析(PARAFAC)作为一种多组分分析数学方法,能够实现多组分混合体系中组成成分的识别和分离,将其与 EEM 结合能够识别 DOM 中的独立荧光组分,并对各荧光组分进行定量.本研究采用文献[14]开发的 DOMFluor 工具箱和 Matlab 2012a 软件对 EEM 数据进行 PARAFAC 分析.

采用平板膜超滤实验考察不同水深处 DOM 膜污染特性.超滤实验在 Amicon 8400 超滤杯(美国,密理博)中进行,采用蠕动泵抽吸的恒流量运行模式,使用的膜通量为 150 $\text{L}\cdot(\text{m}^2\cdot\text{h})^{-1}$,膜过滤实验步骤和膜污染指数的计算见文献[15].

2 结果与讨论

2.1 水库热分层特征

黑河金盆水库 2016 年 7~9 月水体热分层和溶解氧分层状况如图 1 所示.由图 1(a)可以看出,2016 年 7~9 月表层水体与底层水体存在明显温差,水库表层水体在强烈的太阳辐射和大气热交换作用下温度较高,8 月中旬表层水体温度最高达到 28 $^{\circ}\text{C}$;随着水深的增加,太阳辐射迅速衰减且风力混合作用减弱,水深 10~40 m 范围内水温迅速下降;底部水体受太阳辐射和热交换作用影响很小,水温一直保持在 7 $^{\circ}\text{C}$ 左右.7~9 月上下层水体温度差达 15 $^{\circ}\text{C}$ 以上,这导致水体密度随水深增大而增加,水体处于稳定分层状态.

由图 1(b)可以看出,水体热分层期,由于上下水体间物质交换受到抑制,溶解氧也处于分层状态.水库表层溶解氧在 8 $\text{mg}\cdot\text{L}^{-1}$ 以上,基本处于饱和状态;斜温层水体溶解氧基本在 6~7 $\text{mg}\cdot\text{L}^{-1}$ 范围内,低于表层,但随水深增加无明显降低;底部等温层水体溶解氧随水深增加逐渐降低,到 9 月中旬,底部溶解氧已降至 0 $\text{mg}\cdot\text{L}^{-1}$.

2.2 水体分层期 DOM 质量浓度与性质随水深的变化

水体分层期黑河金盆水库不同水深处 DOM 质量浓度如图 2 所示.可以看出,水体分层期 DOM 质量浓度也呈现一定的分层性,斜温层 DOC 质量浓度在 2.69~3.25 $\text{mg}\cdot\text{L}^{-1}$ 范围内变化,高于同期

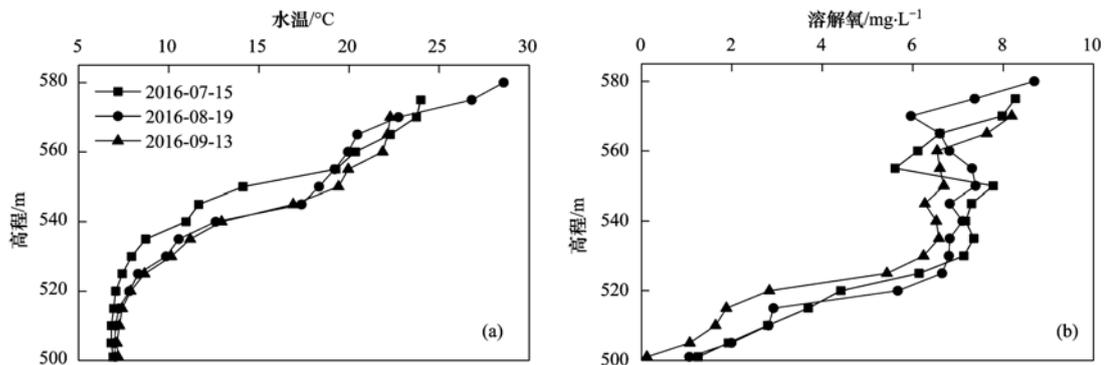


图 1 2016 年 7~9 月黑河金盆水库水体热分层和溶解氧分层特征

Fig. 1 Stratification of the water temperature and dissolved oxygen in the Heihe Jinpen Reservoir during July-Sept. , 2016

等温层和变温层的 DOC 质量浓度. 不同水深处 DOM 质量浓度的差异是外源径流输入、表层水体光化学降解和藻类繁殖、底部沉积-释放等多种过程综合作用的结果. 水体分层期入库径流为等密度潜流, 由于研究期间无强降雨发生, 入库径流泥沙浓度很低, 入流密度主要受水温影响, 可以认为是等温度潜流^[13,16]. 2016 年 7~9 月黑河金盆水库入库径流温度在 14~21℃ 范围内变化, 由图 1(a) 可知, 按照等温度潜流, 分层期入库径流的主要影响范围是水深 15~20 m 处, 因此, 斜温层水体 DOM 质量浓度较高主要是由于径流携带的外源有机物输入造成的. 变温层水体 DOM 质量浓度受光化学降解作用和藻类等浮游植物繁殖影响较大, 太阳辐射造成的光化学降解作用会使部分 DOM 矿化^[17,18], 这是造成变温层 DOM 质量浓度低于斜温层和等温层的重要原因; 而同期的藻密度监测结果表明, 8 月藻类数量出现了小幅度升高, 8 月下旬藻密度达到了 521×10^4 个·L⁻¹, 藻类分泌的有机物可能是造成 8 月变温层 DOM 质量浓度增加的重要原因^[19,20]. 等温层水体 DOM 质量浓度和性质与水-沉积物界面有机物的迁移转化密切相关, 本研究中等温层 DOM 质量浓度低于斜温层, 且在分层期内有所降低, 未出现沉积物中有机物向上覆水体的释

放, 这与黑河金盆水库沉积物较低的污染负荷有关^[21].

紫外吸光度 (UV₂₅₄) 可用于表征含有共轭双键、苯环等不饱和结构的 DOM, 比紫外吸光度 (SUVA) 常用作表征 DOM 芳香化程度的指标, 有研究表明 SUVA 与 DOM 的亲疏水性和分子量大小有一定相关性, 较高的 SUVA 值说明 DOM 中疏水性、大分子组分所占比例较高^[22]. 图 3 为 2016 年 7~9 月黑河金盆水库 DOM 的紫外吸收特征随时间和水深的变化. 可以看出, 水体分层期 DOM 的紫外吸收特性表现出明显的垂向差异, 变温层 DOM 的 UV₂₅₄ 最低 (0.034~0.063 cm⁻¹), SUVA 为 1.52~2.71 L·(mg·m)⁻¹, 这与文献^[23]报道的光化学降解主要破坏 DOM 的不饱和结构以及藻源有机物的芳香度较低相符, 进一步说明了变温层 DOM 主要受光

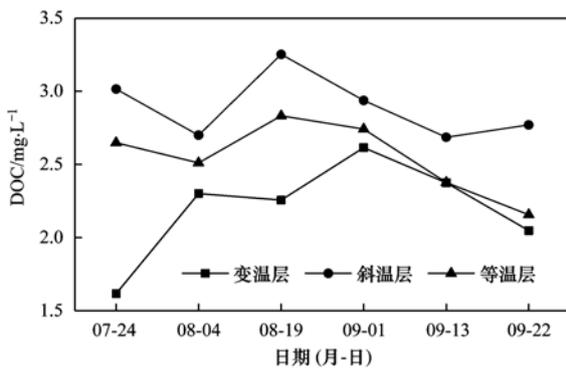


图 2 黑河金盆水库热分层期 DOM 质量浓度的时空变化

Fig. 2 Variation of the DOM concentration in the Heihe Jinpen Reservoir during thermal stratification

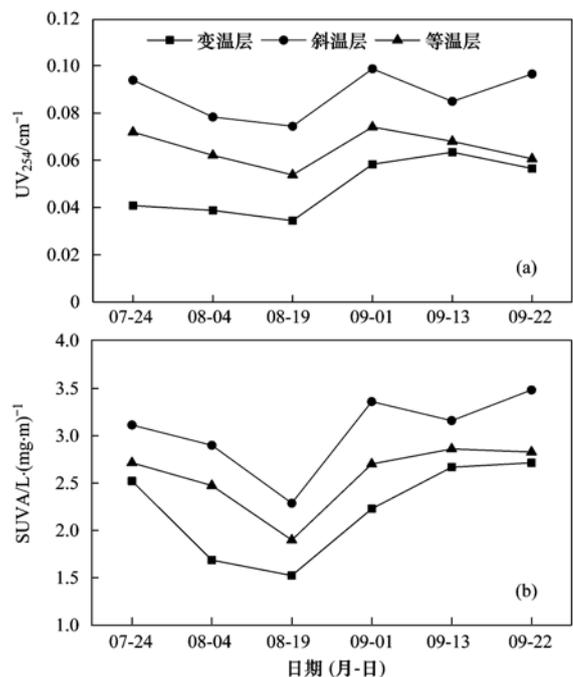


图 3 黑河金盆水库热分层期 DOM 紫外吸收特征的时空变化

Fig. 3 Variation of the DOM's UV absorbance properties in the Heihe Jinpen Reservoir during thermal stratification

化学降解和藻类繁殖影响;斜温层 DOM 的 UV_{254} 最高 ($0.074 \sim 0.099 \text{ cm}^{-1}$), $SUVA$ 为 $2.29 \sim 3.48 \text{ L} \cdot (\text{mg} \cdot \text{m})^{-1}$, 明显高于变温层和等温层 DOM, 这与斜温层水体受入库径流影响较大有关, 黑河金盆水库上游森林覆盖率较高, 入库径流携带的 DOM 主要是芳香度较高的腐殖质类, 因此斜温层 DOM 的芳香度较高^[10].

EEM 光谱灵敏度高, 测定速度快, 样品预处理简单, 近年来已成为研究水中 DOM 性质和迁移转化规律的重要手段. 本研究对热分层期不同水深处 DOM 进行了 EEM 光谱分析, 采用 PARAFAC 分析得到了 3 种荧光组分(图 4), 结合已有报道^[24, 25], C1 组分为类富里酸有机物, C2 组分为类腐殖酸有机物, C3 组分为类色氨酸有机物. 不同荧光组分的来源和特性有较大差异, C1 和 C2 组分的主要来源

为陆源腐殖质, 与 C2 组分相比, C1 组分分子量较小, 疏水性较弱^[26]; C3 组分主要是内源有机物, 一般与藻类、细菌等微生物活动密切相关^[14].

各组分荧光强度随时间和水深的变化如图 5 所示. 可以看出, 变温层中 C1 和 C2 组分荧光强度较低, 说明变温层中的光化学降解作用对腐殖质类的降解作用较强, 这与腐殖质类物质对紫外线较强的吸收能力有关. 斜温层中 C1 和 C2 组分荧光强度高于等温层, 进一步说明入库径流携带的外源腐殖质对斜温层的影响大于对底部等温层的影响. C3 组分与藻源有机物密切相关, 7 月 22 日变温层中 C3 组分荧光强度略高于斜温层和等温层, 8 月 4 日随着表层藻类繁殖增强, 变温层中 C3 组分荧光强度由 7 月 22 日的 0.23 R.U. 增加至 0.49 R.U. , 而斜温层和等温层中 C3 组分仅略有升高, 说明表层水

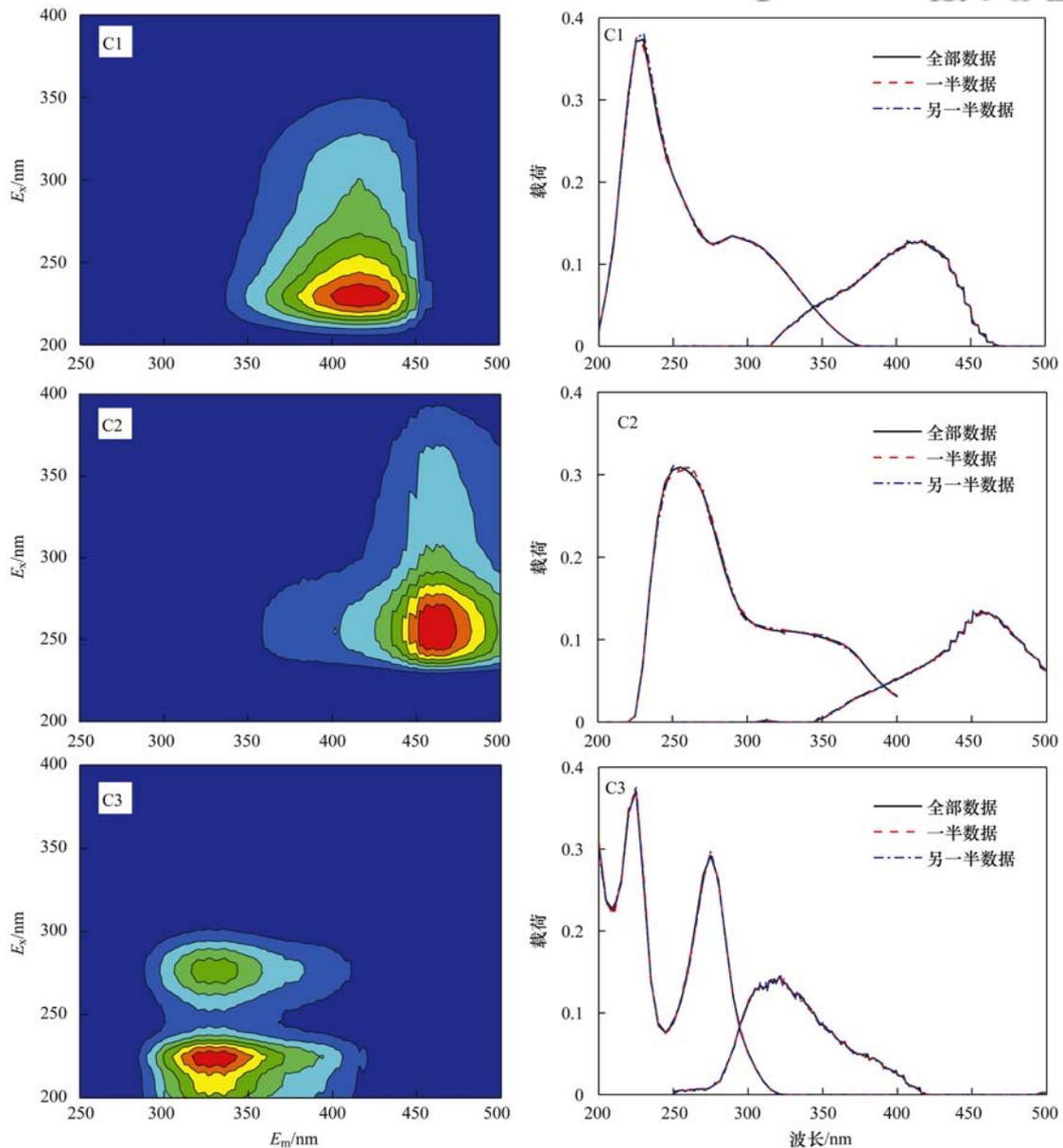


图 4 平行因子分析法解析出的 3 种荧光组分

Fig. 4 Three fluorescence components identified by the PARAFAC analysis

中的藻类繁殖尚未影响到斜温层和等温层；8月19日变温层中C3组分略有下降，而斜温层和等温层中该组分荧光强度有一定增加，这应该是由部分衰老藻细胞沉降以及藻源有机物扩散进入斜温层和等温层水体；之后各层C3组分荧光强度均不断降低，这与C3组分易被生物作用降解有关^[27]。

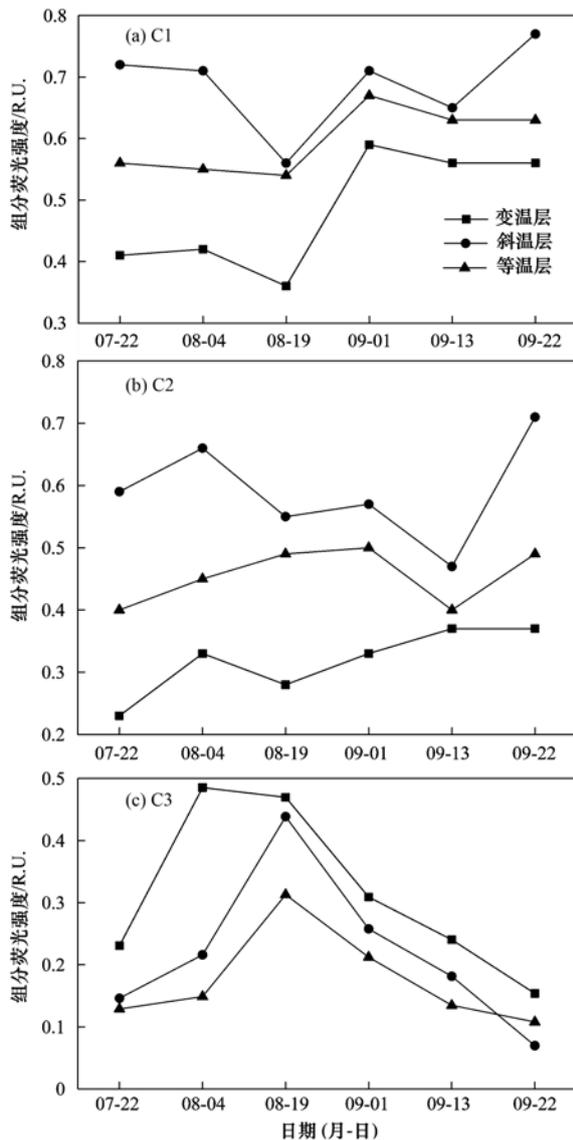


图5 黑河金盆水库热分层期3种组分荧光强度的时空变化
Fig. 5 Variation of the fluorescence intensity of the three components in the Heihe Jinpen Reservoir during thermal stratification

2.3 水体分层期 DOM 的膜污染特性

超滤膜对包括病毒在内的病原微生物具有优异的去除能力，随着超滤膜技术的发展和膜成本的降低，超滤膜在自来水厂中的应用逐渐增加，而 DOM 造成的膜污染是影响超滤工艺推广应用的主要障碍^[28]。为了探明水体分层期 DOM 质量浓度和性质的分层特征对膜污染的影响，使用9月1日采集的水样评价了不同水深处 DOM 的膜污染特征，结果如图6所示。由总污染指数可以看出，变温层 DOM

造成的膜污染最严重，总污染指数为 $(3.42 \pm 0.71) \text{ m}^{-1}$ ，明显高于斜温层和等温层 DOM 造成的膜污染；但水力反洗后剩余的不可逆污染方面，变温层 DOM 造成的不可逆污染指数略低于斜温层和等温层 DOM，这说明变温层 DOM 导致的膜污染的可逆性较好。

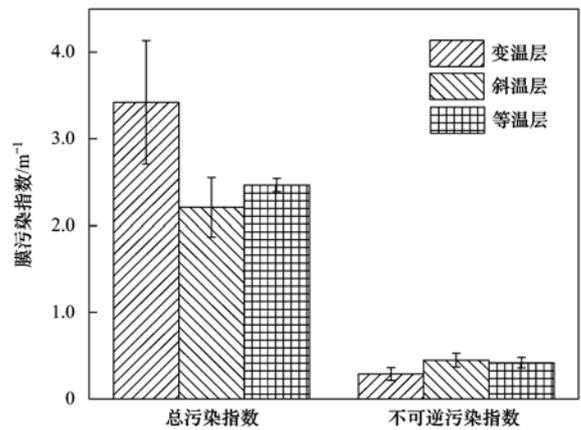


图6 黑河金盆水库热分层期 DOM 膜污染特性
Fig. 6 Fouling potential of DOM in the Heihe Jinpen Reservoir during thermal stratification

为了探究 DOM 组成对膜污染的影响，利用 EEM-PARAFAC 分析得到原水、膜出水、浓缩液、反洗水中各组分的荧光强度，分析了超滤过程中各荧光组分的迁移规律，其中膜上沉积是通过物质平衡得到的反冲洗后仍残留在膜上的部分^[29]，结果如图7所示。可以看出，不同水层 DOM 各荧光组分的迁移规律基本一致。对于 C1 和 C2 组分，膜出水中所占比例为 66% ~ 73%，浓缩液中所占比例为 11% ~ 13%，反洗水中所占比例为 3% ~ 6%，膜上沉积所占比例为 10% ~ 18%；对于 C3 组分，膜出水中所占比例为 53% ~ 59%，浓缩液中所占比例为 8% ~ 10%，反洗水中所占比例为 27% ~ 30%，膜上沉积所占比例为 5% ~ 9%。可以看出，超滤膜对各组分的截留率均高于对 DOM 整体的截留率，说明荧光类 DOM 在膜污染中起着重要作用；超滤膜对 C3 组分的截留率高于 C1 和 C2 组分，说明 C3 组分可能对膜污染有较大影响，这与 C3 组分中分子量较大的蛋白质等生物大分子有关；在反洗水中所占比例方面，C3 组分明显高于 C1 和 C2 组分，这说明被膜截留的 C3 组分易于反洗去除，而被截留的 C1 和 C2 组分很难被水力反洗去除，这可能是由腐殖质类物质较强的疏水性造成的；在膜上沉积所占比例方面，水力反洗之后各荧光组分均有一部分沉积在膜上，说明各荧光组分均造成了不可逆污染，相比而言，C1 和 C2 组分反洗之后沉积在膜上的比例高于 C3 组分。

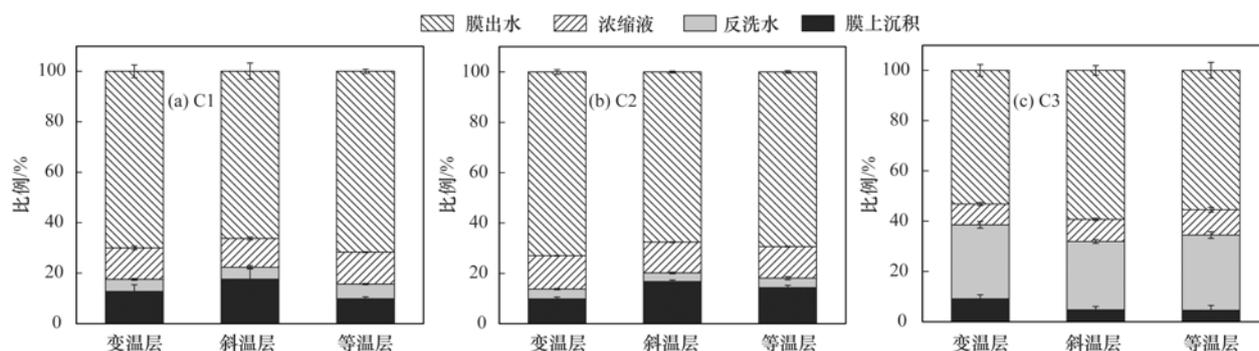


图7 各荧光组分在超滤过程中的迁移

Fig. 7 Migration of the three fluorescence components during ultrafiltration

超滤过程中各荧光组分的迁移规律表明, C3 组分被膜截留的比例较大, 对总污染影响大, 但污染的可逆性较好; C1 和 C2 组分被膜截留的比例小于 C3 组分, 但被截留后难以被反冲洗去除, 对不可逆污染有重要影响. 由于变温层中 C3 组分荧光强度较高, 因此变温层 DOM 造成的总污染最高, 但污染的可逆性较好, 不可逆污染反而略低于斜温层和等温层.

3 结论

(1) 黑河金盆水库夏秋季形成稳定的热分层, 热分层抑制了水体垂向的物质迁移扩散, 使 DOM 的质量浓度和性质也呈现出一定的分层特征.

(2) 变温层 DOM 受光化学降解作用影响较大, 其质量浓度低于中下部水体, 同时受藻类等浮游植物产生的有机物的影响, 芳香度较低, 类富里酸 (C1) 和类腐殖酸 (C2) 组分荧光强度较低, 而类色氨酸 (C3) 组分荧光强度较高.

(3) 在等密度潜流的作用下, 斜温层水体受入库径流影响较大, DOM 质量浓度高于变温层和等温层, 且呈现明显的陆源输入特征, 芳香度较高, C1 和 C2 组分荧光强度较高, C3 组分荧光强度较低.

(4) 与 C1 和 C2 组分相比, C3 组分被超滤膜截留比例较高, 但被截留的 C3 组分易被反洗去除, 可逆性较好. 因此, 虽然变温层 DOM 质量浓度最低, 但由于 C3 组分荧光强度较高, 造成的总污染大于斜温层和等温层, 而不可逆污染小于斜温层和等温层.

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