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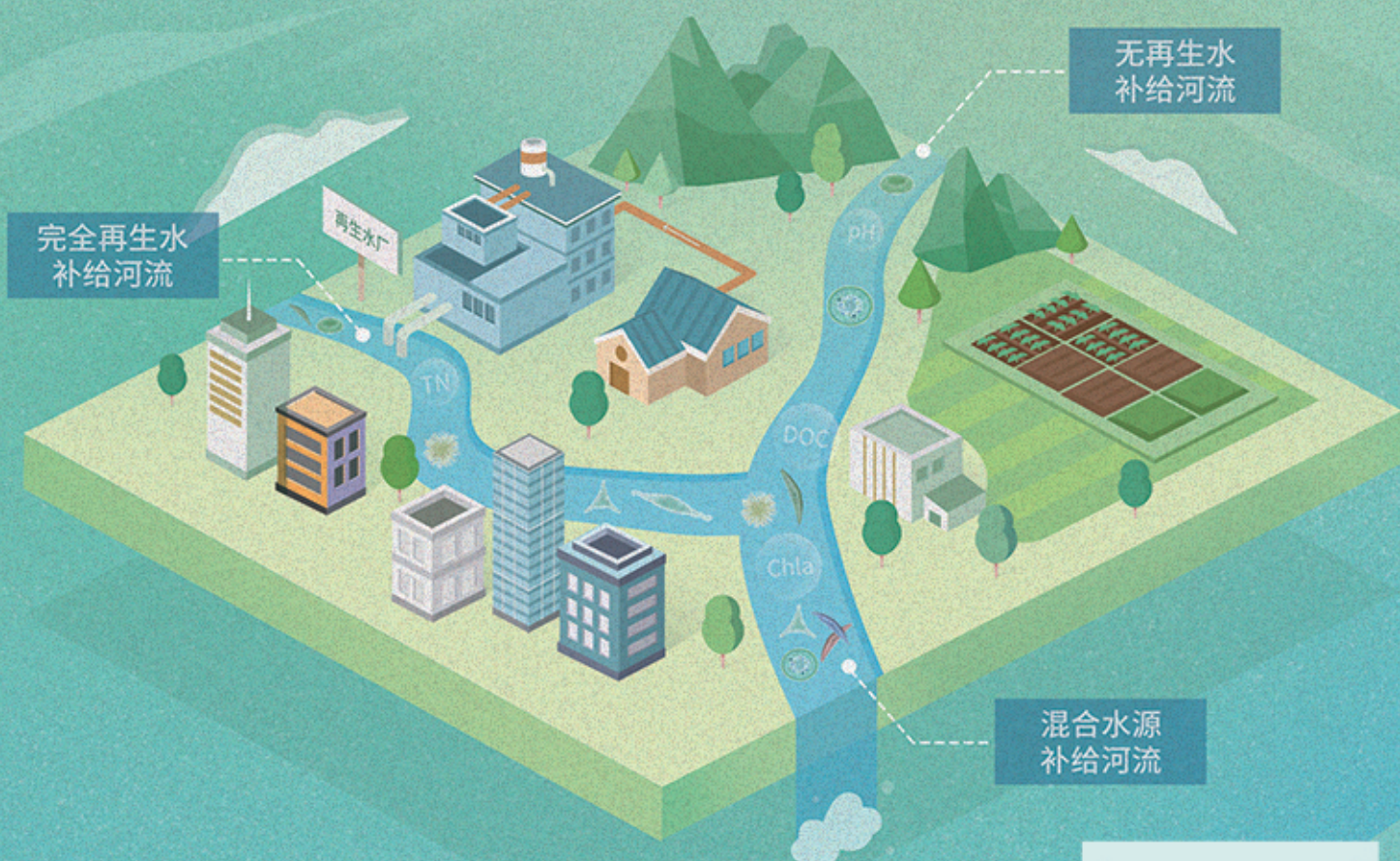
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HUANJING KEXUE

不同水源补给河流浮游植物群落结构特征及其与环境因子的关系

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三峡水库典型消落带土壤 DOM 组分特征及其对有效态镉释放影响

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摘要: 重金属镉(Cd)是三峡水库蓄水后消落带土壤中主要的重金属污染物之一。为研究三峡水库反季节调蓄条件下溶解性有机质(DOM)对有效态Cd在土壤固-液相间释放行为及其再补给过程的影响,以三峡水库香溪河消落带为研究对象,分别于2019年10月和2020年10月采集土壤样品,利用紫外-可见吸收光谱、三维荧光光谱技术和荧光区域积分法分析土壤DOM光谱特性,同时结合梯度扩散薄膜技术(DGT)和土壤扩散通量模型(DIFS)综合探究土壤有效态Cd及其释放动力学过程。此外,测定了消落带土壤基本理化特性以及土壤中Cd含量和赋存形态。结果表明,研究区土壤DOM以富里酸为主,腐殖质特征较弱,具有陆源为主和内源较弱的混合特征,与2019年相比,2020年土壤DOM中富里酸类物质占比更高;2019年和2020年土壤 $\omega(\text{Cd})$ 平均值分别为 $(0.474 \pm 0.301) \text{ mg} \cdot \text{kg}^{-1}$ 和 $(0.249 \pm 0.058) \text{ mg} \cdot \text{kg}^{-1}$,土壤中Cd的赋存形态均以非残渣态为主;DGT测定结果显示2020年土壤中有效态Cd含量较2019年呈下降趋势,进一步通过DIFS模型拟合结果表明,土壤中Cd的吸附速率大于解吸速率,土壤对有效态Cd的再补给能力较弱,表明三峡消落带土壤中有效态Cd的潜在释放风险较低,低分子量DOM和DOM中富里酸类物质是抑制土壤中有效态Cd释放的重要因素。研究结果揭示了三峡水库水文情势变化下DOM对重金属Cd环境地球化学行为的影响,为三峡库区重金属污染防治提供科学理论依据。

关键词: 三峡水库(TGR); 消落带; 溶解性有机质(DOM); 镉(Cd); 梯度扩散薄膜技术(DGT); 迁移释放

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Characteristics of Dissolved Organic Matters and Their Influence on Labile Cadmium Release from Soils of Typical Water Level Fluctuation Zones of Three Gorges Reservoir

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Abstract: Cadmium (Cd) is one of the main heavy metal pollutants in the water level fluctuation zone soils following the impoundment of the Three Gorges Reservoir (TGR). To investigate the influence of dissolved organic matter (DOM) on the release behavior and resupply process of labile-Cd in the soil solid and liquid interphase under the anti-seasonal regulation and storage mode of the TGR, soil samples were collected from the riparian soils along Xiangxi River in October 2019 and October 2020. The UV-visible absorption spectrum, three-dimensional fluorescence spectroscopy, and fluorescence regional integration (FRI) were conducted to analyze the spectral characteristics of DOM. The diffusive gradients in thin films technology (DGT) and DGT-induced fluxes in soils (DIFS) model were used to reveal the labile-Cd and its release kinetics. Further, the soil physical and chemical property, content, and occurrence form of Cd were also determined. The results showed that the DOM was dominated by fulvic acid, and the humification degree was low, the source of DOM was mainly input from the land, and the content of fulvic acid-like materials in the DOM was higher in 2020. The average contents of Cd in soils in 2019 and 2020 were $(0.474 \pm 0.301) \text{ mg} \cdot \text{kg}^{-1}$ and $(0.249 \pm 0.058) \text{ mg} \cdot \text{kg}^{-1}$, respectively, and the non-residual components of Cd were the dominant fraction in the soils. The DGT and DIFS model assay results showed that the content of labile-Cd in soils in 2020 was lower than that in 2019. The adsorption rate of Cd in soils was higher than the desorption rate, the soils had a weak ability to recharge labile-Cd, and the potential release risk of labile-Cd was low, mainly due to the potential inhibition of the release of labile-Cd in soils by low-molecular weight DOM and fulvic acid. These results revealed the influence of DOM on the environmental geochemical behavior of Cd under the change in hydrological situation of the TGR, providing a reference for the management of heavy metals in the TGR area.

Key words: Three Gorges Reservoir (TGR); water level fluctuation zone; dissolved organic matter (DOM); cadmium (Cd); diffusive gradients in thin films (DGT); migration and release

三峡水库因采用“冬蓄夏泄”反季节调蓄模式,形成了垂直距离 30 m,面积达 350 km² 的消落带^[1]。镉(Cd)作为三峡水库消落带土壤中的主要重金属污染物^[2,3],其污染程度处于中度甚至重度水平^[4,5],具有较高的潜在生态风险^[6,7]。然而,以往研究往往基于土壤中Cd的总量评价消落带土壤Cd污染程度和潜在风险,无法客观反映土壤中重金属

Cd的生物有效性^[8,9](即有效态),同时研究采用的传统提取法在揭示Cd在土壤中的迁移释放过程方面也具有一定局限性^[10]。三峡水库特殊的反季节调

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蓄模式打破了库区消落带原有的水土环境^[11], 导致土壤中重金属的环境行为发生改变^[12]. 然而, 消落带水土环境和重金属 Cd 环境行为的变化是否会引起土壤中有有效态 Cd 在土壤固-液两相之间释放行为的改变目前仍不清楚. 梯度扩散薄膜技术(DGT)与土壤扩散通量模型(DIFS)作为揭示土壤重金属有效态及其释放过程的可靠分析技术手段^[13,14], 将二者结合使用, 可以综合探究三峡水库水位变化下土壤有效态 Cd 的变化和其在土壤固-液相间的释放动力学过程, 提升对三峡水库消落带土壤中 Cd 环境地球化学行为的认识.

溶解性有机质(DOM)是重金属重要的载体, 可以与重金属发生吸附、解吸和络合等一系列作用^[15,16]. 有研究表明 DOM 可以与土壤中 Cd 结合, 以 Cd-DOM 络合物形式存在^[17], 促进 Cd 在土壤中的迁移^[18]. Huang 等^[19]通过模拟泄水实验证明土壤中 DOM 强化了 Cd 与土壤的结合, 抑制了 Cd 的解吸过程, 影响其在土壤中的迁移性. 三峡水库消落带“干湿交替”使土壤中 DOM 组分和来源的分布受到影响^[20,21], 这势必会引起土壤中 Cd 的吸附和解吸行为发生变化^[22], 而 DOM 的变化是否影响有效态 Cd 在土壤固-液相中的迁移释放过程目前尚不明晰. 尽管已有研究讨论了 DOM 中的腐殖质组分与 Cd 结合后会降低有效态 Cd 的含量^[23], 但目前仍不清楚三峡水库反季节调蓄模式下 DOM 差异性变化

对土壤固-液相中有有效态 Cd 迁移释放的影响.

本文以三峡水库香溪河消落带为研究对象, 分别于 2019 年 10 月和 2020 年 10 月采集土壤样品, 利用光谱手段并结合 DGT 技术和 DIFS 模型探讨 DOM 组分特征对土壤固-液两相间有效态 Cd 迁移释放过程的影响, 提升对重金属在三峡水库水环境中地球化学循环的理解, 以期为实现三峡库区重金属污染控制和水环境治理提供科学理论依据.

1 材料与方法

1.1 研究区概况与样品采集

研究区为香溪河上游峡口镇至下游河口处, 全长约 15 km. 分别于 2019 年 10 月和 2020 年 10 月, 对研究区上中下游 165 ~ 175 m 处土壤进行采样, 每块区域内均采用五点法取样并将样品混合均匀, 将收集好的土壤样品做好标记后带回实验室风干, 剔除土壤中植物根系和石块等, 研磨过 2 mm 尼龙筛网备用. 采样点分布情况如图 1 所示, 其中 S1 ~ S8 为 2019 年采样点位, S9 ~ S11 为 2020 年采样点位.

1.2 样品理化性质测定

土壤 pH 采用美国 Hach HQ40d 多参数电化学分析仪测定, 水土比为 5:1. 土壤阳离子交换量(CEC)采用氯化钡缓冲溶液法测定^[24]. 土壤样品粒度采用日本 Horiba LA-960A 型激光粒度仪测定. 土壤 DOM 提取方法采用水土振荡提取法^[25], DOM 浓

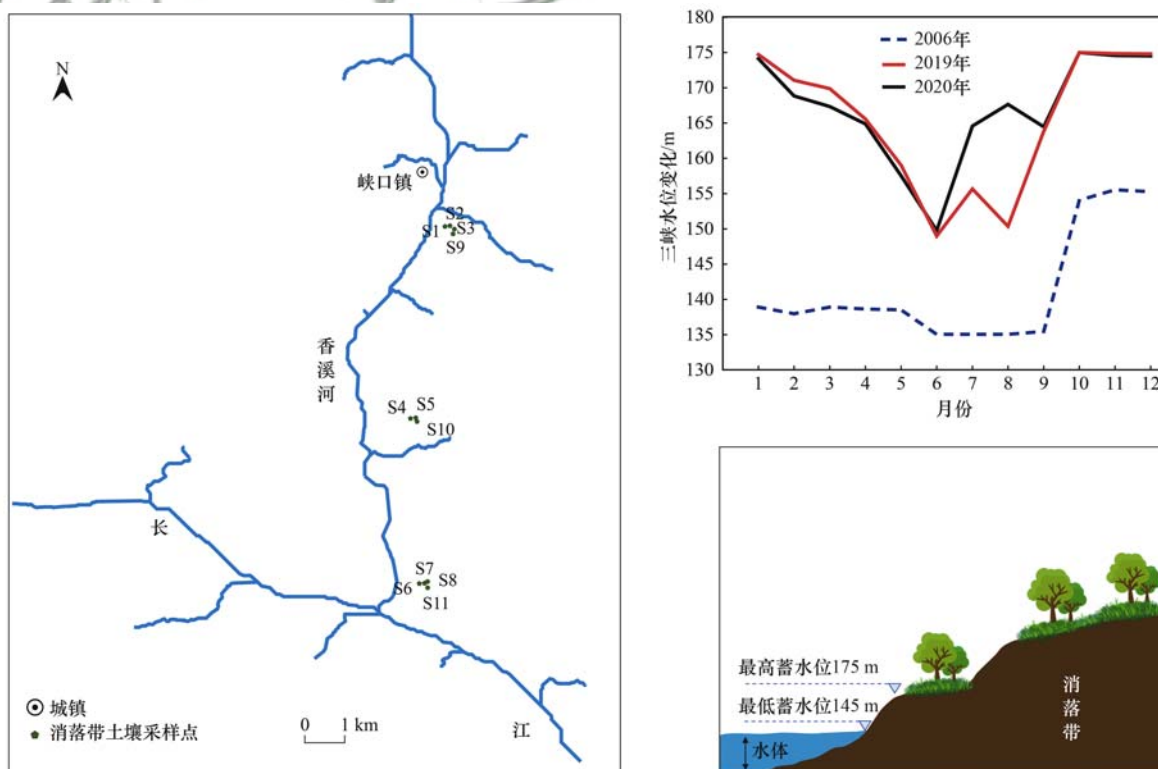


图1 研究区概况和采样点分布示意

Fig. 1 Overview of the study area and sample collection sites

度以溶解性有机碳(DOC)浓度表示,采用德国 vario TOC 分析仪测定 DOC 浓度($\text{mg}\cdot\text{L}^{-1}$)和土壤总有机碳(TOC)含量. DOM 紫外可见吸收光谱采用北京 TU-1810 分光光度计测定,扫描波长为 200 ~ 800 nm,三维荧光光谱采用日本 Hitach F-4600 荧光光谱仪测定,激发波长为 230 ~ 450 nm,发射波长为 250 ~ 620 nm.

土壤固相 Cd 采用硝酸-氢氟酸-双氧水法提取^[26],使用电感耦合等离子体质谱仪(ICP-MS, Agilent 7700x,美国)测定,通过分析空白样品和土壤标准物质(GSS-9)进行质量控制. 土壤液相(孔隙水)中 Cd 采用离心法提取并使用 ICP-MS 测定. 土壤 Cd 不同赋存形态采用改进的欧共体标准物质局(BCR)三步提取法提取^[27],F1、F2、F3 和 F4 依次为酸可提取态、可还原态、可氧化态和残渣态,样品使用 ICP-MS 进行测定,BCR 标准物质(BCR701)用于质量控制,F1、F2 和 F3 的回收率分别为 109.19%、91.51% 和 108.86%. 实验过程中所有玻璃实验器皿均提前用 20% 硝酸溶液浸泡 24 h,实验用水为 Milli-Q 超纯水(电阻率 $18.2\text{ M}\Omega\cdot\text{cm}$),实验试剂均为分析纯.

1.3 土壤重金属有效态浓度的计算及动力学模型

采用标准圆扣式 DGT 装置测定土壤样品中 Cd 的有效态浓度,其计算方法如式(1)和式(2)所示,具体计算参数及操作步骤参考 Gao 等^[28]的研究.

$$M = c_e(V_e + V_g)/f_e \tag{1}$$

$$c_{\text{DGT}} = M \times \Delta g / (D \times A \times t) \tag{2}$$

利用 DIFS 模型模拟 Cd 从土壤固相到液相的释放-再补给动力学过程,计算方法如下^[29]:

$$R = c_{\text{DGT}}/c_{\text{sol}} \tag{3}$$

$$K_{\text{dl}} = \frac{C_s}{c_{\text{sol}}} = \frac{1}{P_c} \frac{k_f}{k_b} \tag{4}$$

$$t_c = 1/(k_f + k_b) \tag{5}$$

式中, R 为重金属有效态浓度(c_{DGT})与液相中浓度

(c_{sol})的比值,反映土壤从固相到液相再补给一种元素的能力,值越大表明再补给能力越强^[30]. K_{dl} 为固液相分配系数($\text{cm}^3\cdot\text{g}^{-1}$); C_s 为固相不稳定 Cd 含量; P_c 为颗粒浓度; t_c 为 Cd 从土壤固相再补给到液相中的响应时间,值越大说明响应时间越长; k_f 和 k_b 分别为吸附速率常数和解吸速率常数(s^{-1}),其数值越高表明土壤吸附或解吸所需要的时间越短.

1.4 数据处理

采用 Excel 2019 和 SPSS 26.0 统计分析软件对原始数据进行相关统计分析,相关图形采用 ArcGIS 10.2 和 Origin Pro 2021 进行绘制.

2 结果与讨论

2.1 消落带土壤基本理化性质及 DOM 组分特征

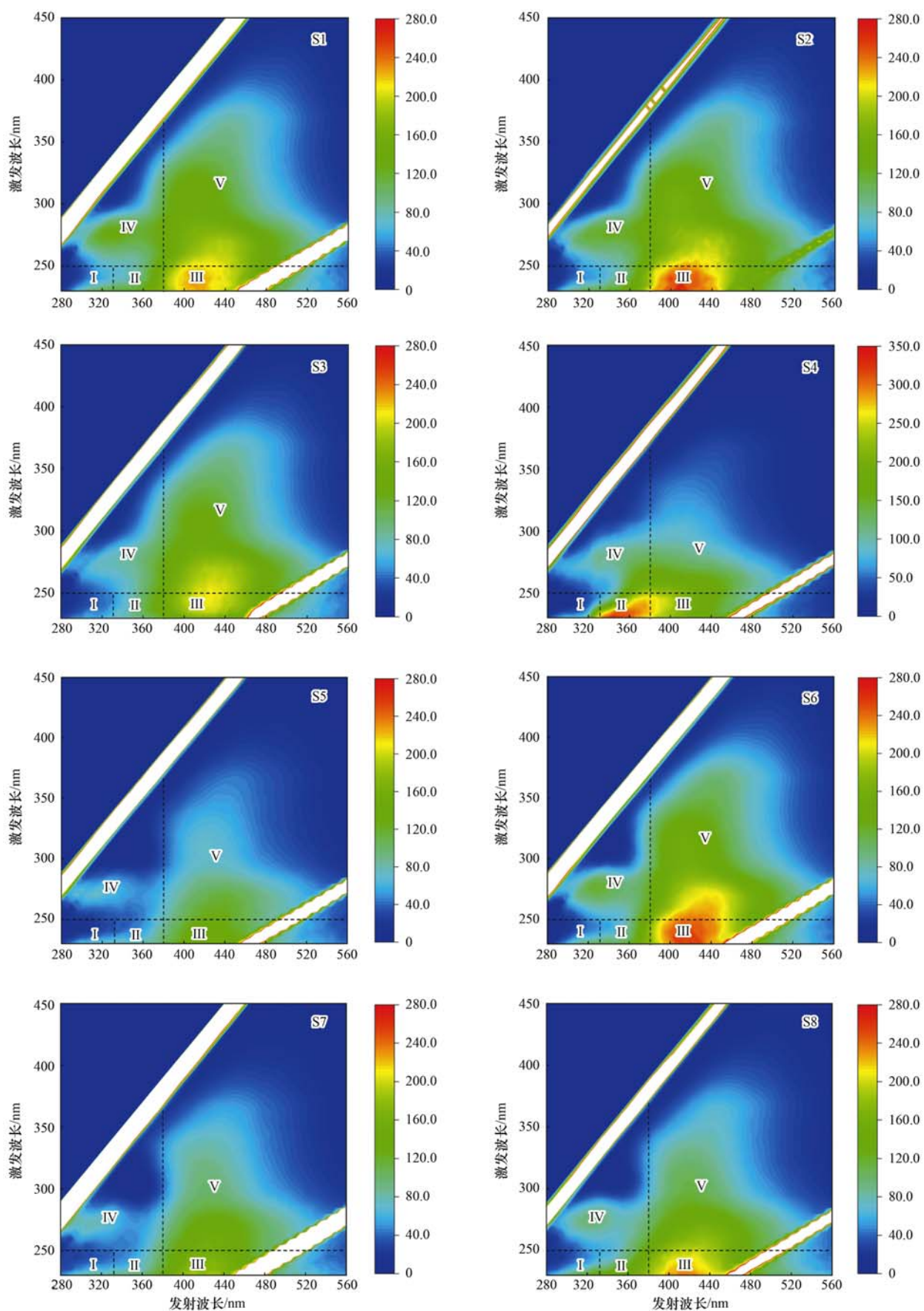
土壤样品基本理化性质如表 1 所示. 研究区土壤 pH 范围在 6.72 ~ 7.95 之间,总体偏中性;土壤中 $\omega(\text{TOC})$ 范围在 2.89 ~ 17.04 $\text{g}\cdot\text{kg}^{-1}$ 之间;土壤粒度组成以砂粒($>20\text{ }\mu\text{m}$)为主;土壤阳离子交换量(CEC)介于 8.87 ~ 13.40 $\text{cmol}\cdot\text{kg}^{-1}$ 之间.

土壤 DOM 特征参数如表 2 所示. $\rho(\text{DOC})$ 范围在 18.2 ~ 27.4 $\text{mg}\cdot\text{L}^{-1}$ 之间,不同位置与 DOM 浓度间无显著性差异($P>0.05$). 结合紫外-可见光谱参数与荧光参数分析可知,所有采样点土壤 DOM 组分均以富里酸为主,来源主要为陆源输入,但夹杂微生物代谢等内源产生的输入;土壤 DOM 的腐殖质特征较弱,自生源特征较显著,生物可利用性较高,该结果与以往的研究结果类似^[25,31]. 各采样点土壤 DOM 的荧光光谱如图 2 所示,利用荧光区域积分法(FRI)对各采样点荧光光谱区域进行划分. 荧光区域积分解析的 5 种组分的分布情况如图 3 所示,土壤 DOM 各荧光区域所占比例中区域 III 所占比例最高,说明土壤 DOM 以富里酸类物质为主,与紫外-可见吸收光谱的分析结果保持一致. 对比 2019 年和

表 1 研究区消落带土壤基本理化参数

Table 1 Soil physical and chemical properties in water-level fluctuation zone

点位	pH	$\omega(\text{黏粒})$ ($<2\text{ }\mu\text{m}$)/%	$\omega(\text{粉粒})$ ($2\sim20\text{ }\mu\text{m}$)/%	$\omega(\text{砂粒})$ ($>20\text{ }\mu\text{m}$)/%	CEC / $\text{cmol}\cdot\text{kg}^{-1}$	$\omega(\text{TOC})$ / $\text{g}\cdot\text{kg}^{-1}$
S1	7.67	1.46	15.92	82.62	12.55	17.04
S2	7.81	0.78	15.94	83.28	11.43	11.88
S3	6.72	0.84	18.50	80.66	12.69	11.53
S4	7.82	1.82	12.28	85.90	9.21	2.89
S5	7.72	1.59	10.40	88.01	10.11	3.50
S6	7.88	0.65	31.36	67.99	8.87	9.48
S7	7.95	2.10	11.63	86.27	11.41	16.36
S8	6.87	1.38	16.73	81.89	11.53	10.72
S9	7.75	0.69	37.51	61.80	13.40	9.58
S10	7.45	1.02	21.07	77.91	12.15	3.97
S11	7.11	0.35	29.36	70.29	9.71	4.49



S1 ~ S11 分别为研究区土壤各采样点编号; 区域 I、II、III、IV 和 V 分别表示酪氨酸类物质、色氨酸类物质、富里酸类物质、微生物代谢产物和腐殖酸类物质

图2 各采样点土壤 DOM 荧光光谱图

Fig. 2 DOM fluorescence spectra of soil at each sampling site

续图 2

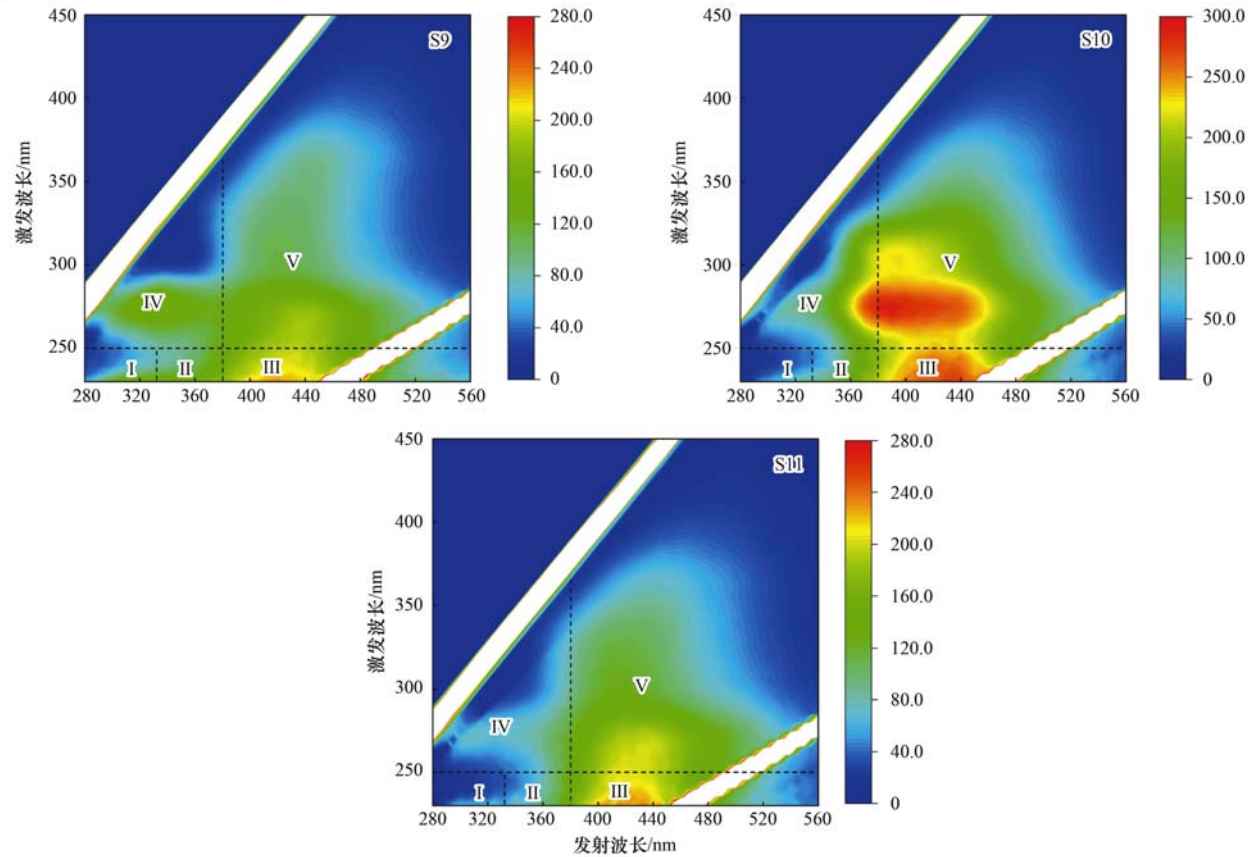


表 2 研究区土壤 DOM 光谱参数

Table 2 Spectral parameters of DOM in soil at each sampling site

点位	$\rho(\text{DOC})$ / $\text{mg}\cdot\text{L}^{-1}$	$a(280)$ / m^{-1}	$a(355)$ / m^{-1}	SUVA_{254} / $\text{L}\cdot(\text{mg}\cdot\text{m})^{-1}$	SUVA_{260} / $\text{L}\cdot(\text{mg}\cdot\text{m})^{-1}$	S_R	FI	HIX	BIX	$\text{Fn}(280)$ /r. u.	$\text{Fn}(355)$ /r. u.
S1	25.3	41.02	19.47	2.09	1.99	0.85	1.57	2.54	0.84	124.56	87.86
S2	24.9	37.43	16.53	1.95	1.87	0.80	1.59	2.79	0.85	121.93	93.93
S3	22.8	35.72	16.36	2.04	1.91	0.95	1.63	3.52	0.72	93.55	87.79
S4	18.2	17.72	7.94	1.32	1.26	0.95	1.64	2.64	0.86	115.23	43.59
S5	20.7	29.46	14.39	1.79	1.70	1.05	1.51	3.34	0.50	52.19	48.76
S6	22.6	31.54	15.13	1.75	1.67	0.79	1.56	3.03	0.58	84.73	69.23
S7	25.2	51.69	24.27	2.60	2.49	0.84	1.57	3.73	0.63	108.96	102.52
S8	27.1	41.64	20.28	2.01	1.93	0.94	1.59	3.15	0.43	55.79	59.24
S9	27.4	50.29	22.98	2.38	2.29	0.97	1.62	2.23	0.54	149.53	89.46
S10	27.4	50.93	23.58	2.38	2.27	0.97	1.56	3.86	1.09	209.70	88.94
S11	26.2	31.91	14.64	1.65	1.57	0.92	1.59	4.57	0.71	80.78	80.87

2020 年土壤 DOM 中组分差异发现,2020 年土壤 DOM 具有更高比例的富里酸类物质.

2.2 土壤 Cd 含量及其各赋存形态分布特征

对 2019 年和 2020 年香溪河消落带土壤固相中 Cd 含量($C_{\text{total-Cd}}$)和液相中 Cd 浓度($c_{\text{sol-Cd}}$)进行了调查,结果如图 4 所示. 2019 年香溪河消落带土壤 $C_{\text{total-Cd}}$ 平均值为 $(0.474 \pm 0.301) \text{mg}\cdot\text{kg}^{-1}$,2020 年为 $(0.249 \pm 0.058) \text{mg}\cdot\text{kg}^{-1}$,均高于三峡库区土壤重金属背景值 $(0.134 \text{mg}\cdot\text{kg}^{-1})$ [32],香溪河沿岸工业废水及生活污水排放可能是造成土壤 $C_{\text{total-Cd}}$ 值高的主要原因 [33,34]. 2019 年和 2020 年土壤固相中 Cd 含量分布呈现相似的规律,下游河口处 Cd 平均含

量高于同年中上游处,这可能与来自香溪河上游的泥沙运移沉积 [35] 和河口水动力条件的变化 [36,37] 有关. $c_{\text{sol-Cd}}$ 范围介于 $0.12 \sim 2.40 \mu\text{g}\cdot\text{L}^{-1}$ 之间,其分布规律和 $C_{\text{total-Cd}}$ 分布规律相似. 土壤 Cd 不同赋存形态占比如图 5 所示,各采样点土壤中 Cd 的赋存形态均以非残渣态 ($F1 + F2 + F3$) 为主,表明香溪河消落带土壤中 Cd 具有一定的迁移性和生物有效性 [38,39].

2.3 土壤有效态 Cd 分布特征及释放动力学过程

研究区土壤有效态 Cd 浓度 ($c_{\text{DGT-Cd}}$) 变化范围介于 $0.02 \sim 0.18 \mu\text{g}\cdot\text{L}^{-1}$ 之间,2019 年平均值为 $(0.08 \pm 0.05) \mu\text{g}\cdot\text{L}^{-1}$,2020 年 $c_{\text{DGT-Cd}}$ 平均值有所下降,为 $(0.05 \pm 0.02) \mu\text{g}\cdot\text{L}^{-1}$. 与丹江口水库和嘉陵

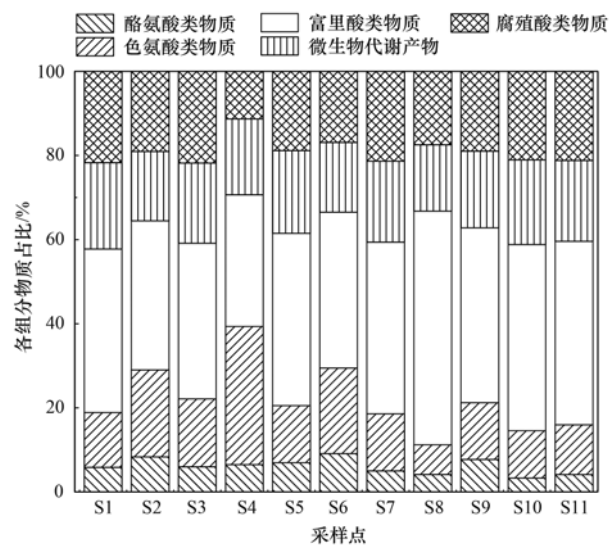


图3 基于荧光区域积分法的土壤 DOM 组分分布
Fig. 3 Distribution of DOM components identified using FRI method in each soil sample

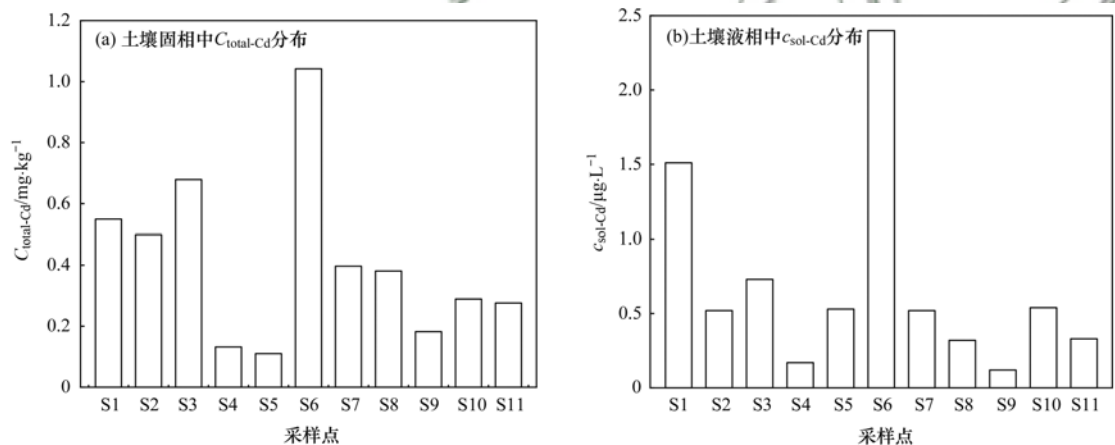


图4 研究区土壤固相及液相中 Cd 分布情况
Fig. 4 Distribution of content of Cd in soil solid and liquid phases in the study area

表3 DIFS 模型模拟结果

Table 3 Simulation results derived from the DIFS model						
点位	$K_{dl}/\text{cm}^3 \cdot \text{g}^{-1}$	R	R_{diff}	t_c/s	k_f/s^{-1}	k_b/s^{-1}
S1	184.05	0.06	0.037	73 650	1.35E-05	3.43E-08
S2	441.98	0.13	0.035	10 190	9.80E-05	9.49E-08
S3	330.51	0.24	0.031	1 567	6.37E-04	6.92E-07
S4	218.39	0.18	0.026	3 465	2.88E-04	3.73E-07
S5	70.45	0.03	0.026	350 800	2.84E-06	1.16E-08
S6	303.05	0.05	0.039	176 200	5.67E-06	9.30E-09
S7	309.83	0.05	0.031	117 500	8.50E-06	9.93E-09
S8	296.59	0.19	0.030	2 590	3.86E-04	4.34E-07
S9	460.00	0.19	0.035	3 889	2.57E-04	2.38E-07
S10	102.03	0.09	0.031	21 430	4.65E-05	1.63E-07
S11	167.27	0.20	0.031	2 411	4.14E-04	8.68E-07

给能力,说明香溪河上游消落带土壤在自然环境变化和人类活动扰动下土壤中 Cd 具有相对较大的潜在释放风险.

江^[40,41]土壤/沉积物中的有效态 Cd 浓度相比,三峡库区消落带土壤中 c_{DGT-Cd} 平均值较低. DIFS 模型模拟结果如表 3 所示,三峡水库香溪河消落带土壤的 R 值均小于 0.25,在时间序列上表现出相似的变化趋势(图 6),即 R 值迅速增加,在 15~30 min 内达到最大值,此后持续呈现下降趋势.该现象说明土壤 Cd 会从土壤固相解吸并释放到土壤液相中,但不能使 Cd 稳定维持在初始状态,并且随着时间推移 Cd 含量会越来越低.此外,同一采样点吸附速率常数(k_f)比解吸速率常数(k_b)高 2~3 个数量级,表明三峡水库消落带土壤中 Cd 从固相解吸的再补给速率远低于土壤液相中 Cd 的耗损速率^[28].与 2019 年相比,2020 年土壤固液相分配系数(K_{dl})更低,说明该年份土壤固相中相对可补给有效态 Cd 的容量较低.对比分析各年份不同区域土壤中有效态 Cd 的释放动力学参数可知,研究区上游消落带土壤较中游和下游具有更大的再补

通过对 c_{DGT-Cd} 与 $C_{total-Cd}$ 和 Cd 不同赋存形态进行线性回归分析发现,土壤 c_{DGT-Cd} 与 $C_{total-Cd}$ 具有显著线性正相关关系($P<0.01$),但二者分布规律并不完

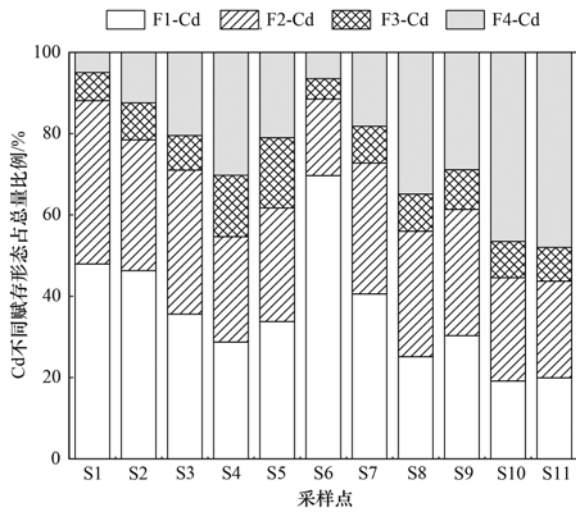
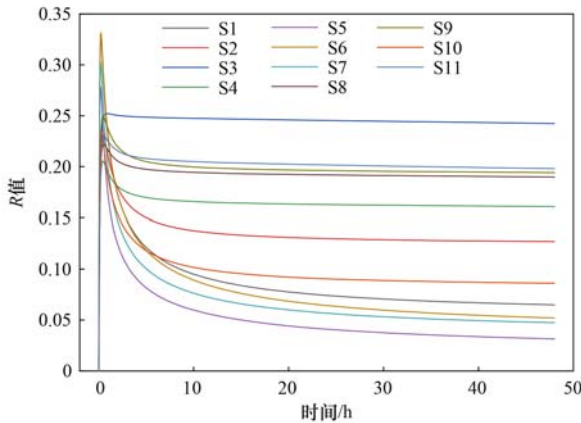


图5 研究区土壤 Cd 不同赋存形态占比

Fig. 5 Proportion of different fractions of Cd in the study area soil

图6 研究区土壤 R 值随时间变化的最佳拟合曲线Fig. 6 Best fitting lines of R values for the study area soil along with the variation time

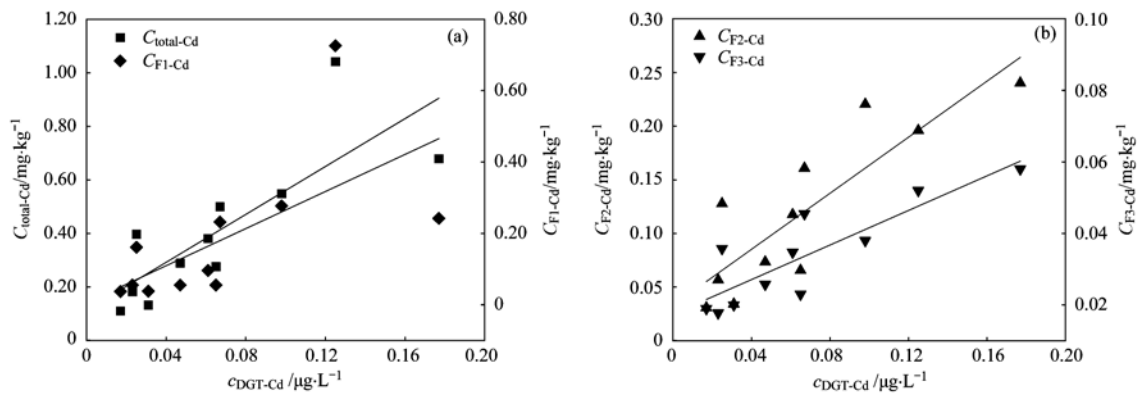
全相同(图7). 点位 S6 处 $C_{\text{total-Cd}}$ 值最高, 但其 $c_{\text{DGT-Cd}}$ 值却不是最高. 此外, $c_{\text{DGT-Cd}}$ 与非残渣态 $C_{\text{F1-Cd}}$ 、 $C_{\text{F2-Cd}}$ 和 $C_{\text{F3-Cd}}$ 之间也具有显著相关性 ($P < 0.01$), 该结果说明土壤中一些物质如有机质、铁/锰氧化物和硫化物等会吸附 Cd 或与之结合^[38,42], 形成不稳定结

合物, 影响 Cd 在土壤固-液相间的释放. 在环境变化和人类活动影响下, 又会再次将部分 Cd 从土壤固相中释放到液相中, 使 $c_{\text{DGT-Cd}}$ 值增加, 导致消落带土壤环境的二次污染.

总体而言, 香溪河消落带土壤对 Cd 的再补给能力较弱, 土壤中有机质、铁/锰氧化物或硫化物等会与 Cd 发生吸附、络合等一系列反应, 影响有效态 Cd 的浓度分布及释放行为. 此外, 各项结果表明三峡水库消落带土壤固相释放-再补给 Cd 的能力较低, 以往基于土壤 Cd 总量^[4,7] 或赋存形态^[43] 的评价方法可能会高估三峡水库消落带土壤 Cd 的潜在生态风险.

2.4 土壤 DOM 对有效态 Cd 释放-再补给过程的影响

土壤 DOM 中不同组分对 Cd 在土壤中吸附解吸行为影响具有差异性^[44,45]. 主成分分析 (PCA) 显示 DOM 中类蛋白质物质和富里酸类物质是影响有效态 Cd 释放的重要参数指标 (表 4), 进一步通过 Pearson 相关性分析发现, $c_{\text{DGT-Cd}}$ 值和 DIFS 模型模拟计算的 R_{diff} 值与土壤 DOM 中富里酸类物质之间存在一定程度的负相关关系, 与类蛋白质物质之间具有正相关关系, 该结果表明 DOM 中富里酸类物质对有效态 Cd 在固-液相中的释放存在潜在抑制作用. 有研究表明, 富里酸类物质和类蛋白质物质中的羧基和酚羟基类含氧基团对金属结合能力会产生影响^[46]. Zhang 等^[47] 也指出, 富里酸类物质与 Cd 的相互作用比类蛋白更稳定. 土壤中 Cd 与富里酸类物质中主要活性基团 (如羧基和酚基等) 结合的稳定性和类蛋白质物质中阳离子基团的结合稳定性更强^[48], 这也限制了 Cd 从土壤固相中的解吸释放行为. 2020 年三峡水库消落带土壤 DOM 具有较高比例的富里酸类物质 (图 8), 这表示三峡水库消落带土壤经一年水位涨落调度后, 土壤 DOM 中富里酸类物质占比有所增加, 而类蛋白质物质占比相对下降;

(a) $c_{\text{DGT-Cd}}$ 与 $C_{\text{total-Cd}}$ 和 $C_{\text{F1-Cd}}$ 之间的关系, (b) $c_{\text{DGT-Cd}}$ 与 $C_{\text{F2-Cd}}$ 和 $C_{\text{F3-Cd}}$ 之间的关系图7 研究区土壤 $c_{\text{DGT-Cd}}$ 与 $C_{\text{total-Cd}}$ 和 Cd 不同赋存形态之间的关系Fig. 7 Relationship among $c_{\text{DGT-Cd}}$, $C_{\text{total-Cd}}$, and the different fractions of Cd

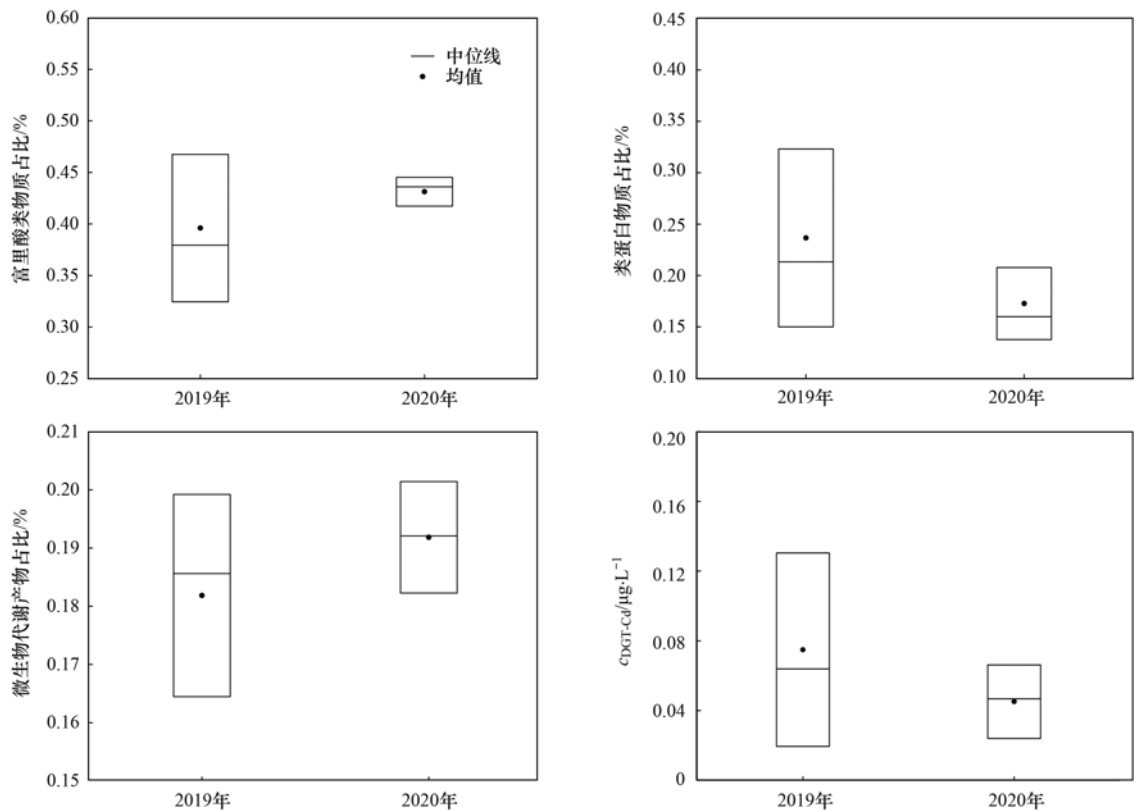


图 8 不同年份土壤 DOM 组分占比和土壤有效态 Cd 浓度变化

Fig. 8 Changes in DOM component proportion and c_{DGT-Cd} of the riparian soil in different years

在富里酸类物质大量存在的情况下,土壤中的 Cd 更容易被吸附而非解吸,抑制了有效态 Cd 从土壤固相到液相间的迁移释放行为,从而导致库区土壤有效态 Cd 浓度下降.

表 4 有效态 Cd 释放的主成分分析结果

Table 4 Principal component analysis results of labile-Cd release			
主成分因子	特征值	差异百分比/%	累积百分比/%
酪氨酸类物质	7.417	33.72	33.72
色氨酸类物质	4.465	20.29	54.01
富里酸类物质	3.873	17.60	71.61
微生物代谢产物	2.640	12.00	83.62
腐殖酸类物质	1.419	6.45	90.06
R_{diff}	1.049	4.77	94.83
k_f	0.592	2.69	97.52
k_b	0.303	1.38	98.90

光谱斜率比值 S_R 被用来指示 DOM 相对分子质量, S_R 越大表明 DOM 相对分子质量越小^[49,50]. Pearson 相关性分析显示 S_R 与 R_{diff} 值之间具有负相关性($r = -0.733$, $P < 0.05$), 并与土壤中 Cd 的总量及赋存形态具有显著负相关关系($P < 0.01$), 和 c_{DGT-Cd} 之间存在负相关趋势, 推测 DOM 分子量大小会影响有效态 Cd 的释放行为. 有研究表明, DOM 与 Cd 的结合能力会随其分子量降低而呈现增加趋势^[51,52]. 2020 年土壤 DOM 的 S_R 值略大于 2019 年, 表明 2020 年香溪河消落带土壤 DOM 相对分子质量

略小于 2019 年, DOM 与 Cd 的结合较为稳固, 土壤在受到环境扰动如 pH 或含水率变化后难以再次释放有效态 Cd, 导致 2020 年土壤中有有效态 Cd 浓度低于 2019 年; 此外, 低分子量 DOM 与 Cd 结合也会导致土壤固相中不稳定 Cd 的含量减少, 在一定程度上限制了有效态 Cd 来自土壤固相中的再补给, 抑制有效态 Cd 的释放, 进而影响有效态 Cd 的浓度大小. 因此, DOM 分子量是影响土壤中有有效态 Cd 释放的重要因素, 且低分子量 DOM 对该过程具有潜在抑制作用.

3 结论

- (1) 三峡水库香溪河消落带土壤 DOM 主要为富里酸, 来源以陆源输入为主, 并夹杂微生物代谢等内源输入. 土壤 DOM 腐殖质特征较弱, 自生源特征较显著, 生物活性较强. 2020 年消落带土壤 DOM 较 2019 年具有更高的富里酸类物质和更低的类蛋白质占比.
- (2) 2019 年和 2020 年香溪河消落带土壤 Cd 含量均高于三峡库区土壤重金属背景值, 土壤液相中 Cd 浓度与土壤 Cd 总量分布规律相似, 土壤中 Cd 主要赋存形态为非残渣态 ($F1 + F2 + F3$).
- (3) 2020 年土壤有效态 Cd 浓度低于 2019 年, 土壤固相中 Cd 的吸附速率大于解吸速率, 土壤对

有效态 Cd 从固相至液相的再补给能力较弱,表明香溪河消落带土壤有效态 Cd 的潜在释放风险较低。

(4)香溪河消落带土壤 DOM 中的富里酸类物质和土壤中低分子量 DOM 均潜在地抑制了有效态 Cd 在土壤固-液相中的释放-再补给动力学过程。

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