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三峡支流消落带表层沉积物氮矿化动力学参数估算

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摘 要:为揭示干湿循环过程对消落带沉积物氮矿化动力学特征的影响,以三峡典型支流——澎溪河为例,分别于上、下游两个水文断面采集 170 m(高)和 150 m(低)水位高程消落带的表层(0~15 cm)沉积物样品。采用厌氧培养法测定其净氮矿化速率,应用 One-pool、Two-pool、Special 和有效积温(EATM)四种模型拟合氮矿化过程并建立估算方程。结果表明,沉积物总碳、总氮、碳氮比、有机质、粘粒、粉粒和铵态氮含量随高程降低而增加,而沉积物总磷、砂砾、硝态氮趋势相反($P<0.05$)。低、高水位高程氮矿化最优拟合模型分别为 One-pool 和 Special,其中易矿化速率常数(k_d)分别为 0.17、0.12 d⁻¹,易矿化势(f_d)分别为 4.05%和 4.71%。另外, f_d 与总碳、总氮、碳氮比、有机质、铵态氮、粉粒呈显著负相关,与硝态氮和砂砾呈显著正相关($P<0.05$),而 k_d 相反,且两参数可用有机质和碳氮比进行估算。

关键词:三峡库区;氮矿化速率;水位波动;氮矿化模型;澎溪河流域

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Parameter estimates of sediment nitrogen mineralization kinetics in the water level fluctuation zone of a Three Gorges Reservoir tributary

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Abstract: To reveal the effect of drought–rewetting process on the nitrogen mineralization kinetics in the sediment of the water level fluctuation (WLF) zone, surface sediment samples were collected over various water level altitudes at the upper and downstream sections of the Pengxi River. The sediment nitrogen mineralization rate was evaluated with anaerobic incubation. The one–pool, two–pool, special, and effective accumulated temperature models (EATM) were applied to fit the data of nitrogen mineralization kinetics in sediments. The result showed that total organic carbon (C), total nitrogen (N), organic matter (OM), clay, silt, and $\text{NH}_4^+\text{--N}$ in the higher altitude were lower than that in the lower altitude, which is on the contrary to total phosphorus, sand, and $\text{NO}_3^-\text{--N}$. The one–pool model and special model were the optimal models for fitting nitrogen mineralization in sediments at low and high altitudes, respectively. The N mineralization rate constants (k_d) were 0.17 d⁻¹ and 0.12 d⁻¹, while the N mineralization potentials (f_d) were 4.05% and 4.71% in the low and high altitude sediments, respectively. The f_d was negatively correlated with C, N, C/N ratio, OM, $\text{NH}_4^+\text{--N}$, and silt, and was positively correlated with $\text{NO}_3^-\text{--N}$ and sand

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($P < 0.05$). The k_d value had an opposite trend compared to the variation in f_d . The model parameters of f_d and k_d were successfully estimated by using the OM and C/N ratio. The current study revealed that nitrogen mineralization kinetics in sediment had a strong correlation with the water nitrogen content, and could thus be used to predict the contribution of nitrogen input into the WLF zone as an indicator of river eutrophication.

Keywords: Three Gorges Reservoir area; nitrogen mineralization rate; water level fluctuation; nitrogen mineralization model; Pengxi River basin

土壤氮素主要以有机氮形态存在,是无机氮的重要来源,其矿化特征对植物养分供给具有重要意义,但其需经微生物转化成铵态氮和硝态氮才能被植物直接利用^[1]。而无机氮极易淋溶进入水体,从而增加水体富营养化风险。

特殊的调蓄水制度使三峡支流不同水位高程消落带经历了不同程度的干湿循环^[2-3],消落带生源要素周转速率、氧化还原状态、水土界面过程等均发生了明显变化^[4-5]。三峡支流蓄水期水流缓慢^[6],氮素进入水体不易迁移扩散,可能造成水体富营养化等水生态问题,因此,查明干湿循环条件下,三峡支流消落带沉积物氮矿化动力学过程至关重要。

氮矿化是陆地生态系统氮循环的关键^[7],主要受凋落物输入、微生物和酶活性以及根际过程等生物因子^[8-10],以及温度、湿度、pH等非生物因素的影响^[11-14]。氮矿化动力学参数可用来衡量氮矿化潜力和供氮能力,通过模型拟合可进行动力学参数估算。氮矿化模型按有机物分解过程可分为零阶、一阶、双一阶和混阶动力学模型^[15-17];按建模方式可分为有效积温模型(EATM)、机理模型和功能模型^[18];按有机氮性质分为One-pool、Two-pool、Special及多氮库模型^[19]。张玉玲等^[20]研究表明,Special模型是长期施肥水稻土氮矿化过程的最佳模型;卢红玲等^[21]研究黄土高原石灰性土壤氮矿化模型发现,Two-pool和Special模型优于One-pool和有效积温模型,且Special模型淹水条件下更优;Gil等^[22]研究发现,Special模型更适合长期施肥后土壤氮矿化过程模拟;Li等^[23]研究上海地区水稻土壤氮矿化模型发现,Two-pool和Special模型对氮矿化的过程模拟效果最好,且Special模型的参数最优;Camargo等^[24]对巴西南部土壤氮矿化过程进行模型拟合,发现One-pool和Two-pool模型拟合效果较好,但参数估算过程较为复杂;刘青丽等^[25]研究表明,在变温条件下有效积温模型能更好地模拟土壤氮矿化过程,而指数模型能较好地描述氮矿化对水分变化的响应。可见,查明不同环境条件下氮矿化动力学最佳模型至关重要。

本研究以三峡支流彭溪河消落带为研究对象,从有机氮分解过程角度结合One-pool、Two-pool、Special、EATM对消落带沉积物氮矿化过程进行了拟合,通过多元回归建立了基本理化性质与拟合参数的估算方程,旨在为查明三峡支流水体富营养化频发和消落带植被适生性下降等问题提供科学依据。

1 材料与方法

1.1 研究区域与样品采集

三峡库区特殊的调蓄水制度使得库区水位在145~175 m之间呈年际周期性涨落。本研究于2015年6月库区水位最低、消落带裸露期间采集三峡支流彭溪河上游(渠口镇)和下游(双江镇)两个水文断面的低水位(150 m)和高水位(170 m)高程的表层(0~15 cm)沉积物样品,每个水位高程由3个随机采样点组成(图1)。采集的原状新鲜样品于4℃保存,一部分用于氮矿化培养实验,另一部分经冻干、剔除植物根系、过筛后用于基本理化性质测定。

1.2 水文特征

2013—2017年三峡库区万州水文站水位波动和水位高程与淹水时间的关系见图2。消落带水位高程和淹水时间呈显著负相关,水位越低淹水时间越长,150 m水位高程淹水时间约325 d·a⁻¹,主要集中在8月至次年6月,170 m水位高程主要淹水时间约123 d·a⁻¹,主要集中10月至次年1月。

1.3 氮矿化实验

采用连续淹水厌氧培养法对氮矿化速率进行了测定^[26],具体步骤如下:准确称取10.00 g经预处理样品于50 mL培养瓶,按水土比2:1加入去离子水,以高纯氮(>99.99%)保持厌氧,控制氧气<1×10⁻⁶,使体系始终处于厌氧状态,密封,每个样品3次重复,于35℃恒温培养箱中避光培养^[27]。分别于第3、7、14、21、28 d破坏性取样,因厌氧条件,故不考虑NO₃⁻-N变化,只测定NH₄⁺-N含量^[28]。

1.4 分析方法

pH值采用0.01 mol·L⁻¹ CaCl₂浸提法测定,有机

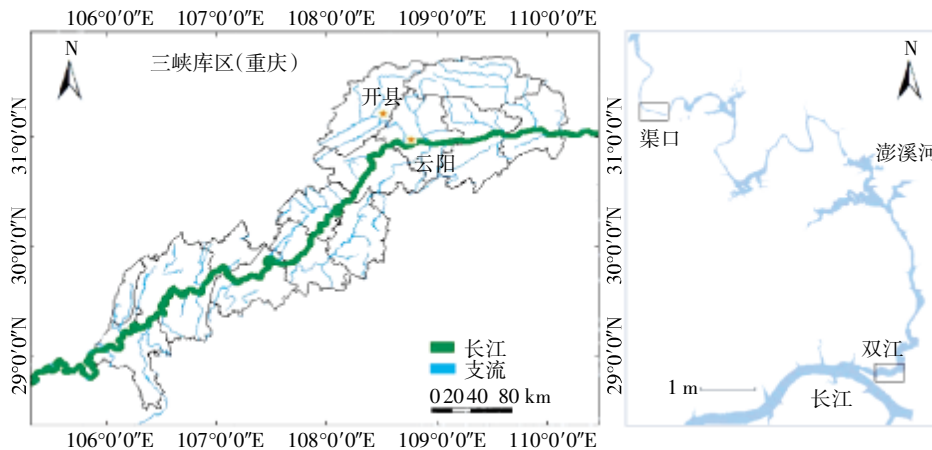


图1 澎溪河消落带采样点位

Figure 1 Sampling sites in the water level fluctuating zone of Pengxi River

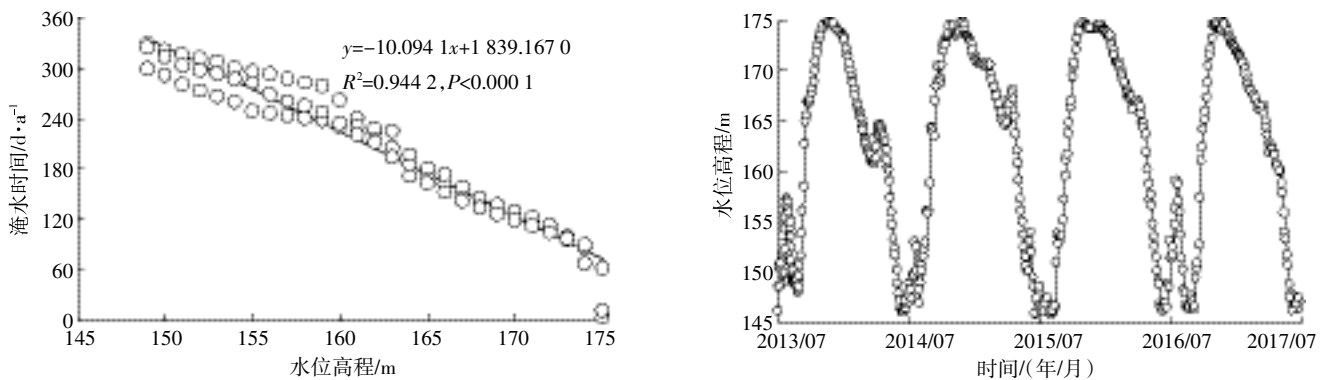


图2 水位高程与淹水时间之间的关系

Figure 2 Relationship of water level altitude with flood time

质(OM)采用重铬酸钾容量法测定, $\text{NH}_4\text{-N}$ 采用靛酚蓝比色法测定, $\text{NO}_3\text{-N}$ 采用 $2\text{ mol}\cdot\text{L}^{-1}$ KCl 浸提比色法测定, 沉积物粒径组成采用比重法测定, 总磷(P)采用碱熔-钼锑抗分光光度法测定, 总碳(C)和总氮(N)用元素分析仪(意大利 EA3000)测定, 溶解性有机碳(DOC)用总有机碳分析仪测定(TOC-VC PN)。

1.5 氮矿化模型

1.5.1 One-pool 模型

One-pool 模型是在一阶指数模型基础上提出的, 假设氮库由单一组分组成, 具体如下^[29]:

$$N_m = N \times 10^3 \times f_d \times (1 - e^{-k_d t}) \quad (1)$$

式中: N_m 为累积氮矿化量, $\text{mg}\cdot\text{kg}^{-1}$; N 为总氮含量, $\text{g}\cdot\text{kg}^{-1}$; f_d 为易矿化氮占总氮比值, %; k_d 为易矿化氮矿化速率常数, d^{-1} 。

1.5.2 Two-pool 模型

将有机氮库分为两部分, 即易矿化氮库和难矿化氮库, 具体如下^[30]:

$$N_m = N \times 10^3 \times f_d \times (1 - e^{-k_d t}) + N \times 10^3 \times (1 - f_d) \times (1 - e^{-k_r t}) \quad (2)$$

式中: k_r 为难矿化氮矿化速率常数, d^{-1} 。

1.5.3 Special 模型

Special 模型是在双库氮矿化模型基础上提出的, 假设氮库存在一个较稳定且较慢矿化的部分, 且该部分更符合零阶方程, 具体如下^[31]:

$$N_m = k_i + N \times 10^3 \times f_d \times (1 - e^{-k_d t}) \quad (3)$$

式中: k_i 为较慢矿化部分矿化速率常数, d^{-1} 。

1.5.4 有效积温模型(EATM)

有效积温模型是以温度为主导因素的模型, 具体如下^[32]:

$$N_m = k \times [(T - T_0) \times t]^n \quad (4)$$

式中: T 为培养温度, $^{\circ}\text{C}$; T_0 为基点温度, $^{\circ}\text{C}$; t 为培养时间, d ; k 和 n 为矿化常数。 $n < 1$ 时, 单位有效积温所矿化氮量随培养时间的增加逐渐减少; $n > 1$ 时相反。

1.6 模型评价与分析

利用 Microsoft Excel 2010 对数据进行处理, 利用

SigmaPlot 12.0 对四种氮库模型进行拟合及绘图,利用 IBM SPSS Statistic 20 对数据进行统计分析,并通过多元逐步回归分析建立基于沉积物基本理化性质的氮矿化动力学参数估测方程,用调整的确定系数和均方根误差判断模型优劣。

模型调整的确定系数(R_{adj}^2)公式为:

$$R_{adj}^2 = 1 - (1 - R^2) \times \frac{n-1}{n-M-1} \quad (5)$$

式中: n 指观测样品数; R^2 为模型确定系数; M 为变量个数,理论上 R_{adj}^2 位于 0~1 之间,其越接近 1,表明模型模拟越准确。

均方根误差(RMSE)公式为:

$$RMSE = \sqrt{\frac{\sum_{i=1}^n (y_{m,i} - y_{p,i})^2}{n}} \quad (6)$$

式中: y_m 指观测值; y_p 指估测值。RMSE 越小,表明预测误差越小,模型精度越高。

2 结果与分析

2.1 消落带沉积物基本理化性质

沉积物基本理化性质见表 1。从表 1 可知,研究区沉积物 C、N、C/N、OM 在低水位高程含量更高,而 P 与之相反,DOC 在水位高程分布上无显著差异。沉积物粘粒和粉粒在水位高程上分布表现为低水位高程>高水位高程,而砂砾与之相反。总体上,砂砾>粉粒>粘粒。 NH_4^+-N 表现为低水位高程>高水位高程,而 $NO_3^- - N$ 分布与其相反。

2.2 氮矿化动力学模型拟合

对三峡支流消落带沉积物氮矿化过程采用 One-

pool、Two-pool、Special 及有效积温模型进行拟合见图 3,模型参数见表 2。在水位高程上,净氮矿化累积量均表现为低水位高程大于高水位高程,且随时间延长显著增加($P<0.05$);有效积温模型对不同水位高程沉积物矿化情况进行拟合得到的 n 值均小于 1,表明单位有效积温所矿化氮量随培养时间的增加逐渐减少。 k 值代表矿化强度,相关分析表明, k 值与累积矿化氮呈显著正相关。One-pool 模型在低水位高程 N_d 值最大, RMSE 值最低, Special 模型在高水位高程 N_d 值最大, RMSE 值最低。

2.3 沉积物动力学参数与基本理化性质相关性

将氮矿化模型拟合参数与沉积物基本理化性质进行相关分析(表 3),结果表明, f_d 与 C、N、C/N、 NH_4^+-N 、OM、粉粒呈极显著负相关($P<0.01$),与砂砾、 $NO_3^- - N$ 呈极显著正相关($P<0.01$); k_d 与 C、N、C/N、 NH_4^+-N 、OM、粉粒呈极显著正相关($P<0.01$),与砂砾、 $NO_3^- - N$ 呈极显著负相关($P<0.01$)。可见, C、N、C/N、OM、 NH_4^+-N 、 $NO_3^- - N$ 、粉粒和砂砾可能为预测沉积物氮矿化动力学参数的决定因子。

2.4 氮矿化模型参数预测方程

将模型参数 f_d 和 k_d 作为因变量,利用相关性分析所得预测沉积物氮矿化动力学参数的决定因子(C、N、C/N、OM、 NH_4^+-N 、 $NO_3^- - N$ 、粉粒和砂砾)作为自变量进行多元逐步线性回归,建立的模型参数预测方程见表 4。从表 4 可知,氮矿化动力学模型参数 f_d 和 k_d 可用 C/N 和 OM 进行估算,模型参数 f_d 和 k_d 的 R_{adj}^2 分别为 0.985 和 0.963, RMSE 分别为 0.006 和 0.000 4, P 值均小于 0.01。可见,该预测方程可较好地预测消落带氮矿化模型参数 f_d 和 k_d 。

表 1 消落带沉积物基本理化性质

Table 1 Physico-chemical properties of the sediments in the WLF zone

水位高程	项目	pH	C/g·kg ⁻¹	N/g·kg ⁻¹	C/N	P/g·kg ⁻¹	NH_4^+-N /mg·kg ⁻¹	$NO_3^- - N$ /mg·kg ⁻¹	OM/%	DOC/mg·kg ⁻¹	粘粒/%	粉粒/%	砂砾/%
低水位	极小值	6.70	18.61	0.93	19.76	0.25	8.71	19.32	1.66	52.64	6.70	21.9	63.05
	极大值	7.60	19.49	0.99	19.96	0.29	11.03	24.06	1.73	58.27	13.55	27.0	66.30
	均值	7.27	19.16	0.97	19.86	0.27	9.64	22.46	1.70	55.03	10.68	24.1	65.22
	标准差	0.49	0.48	0.03	0.10	0.02	1.23	2.72	0.04	2.91	3.56	2.62	1.88
	偏度	-1.65	-1.59	-1.45	0.30	-0.02	1.46	-1.73	-0.73	1.21	-1.27	1.12	-1.73
	变异系数/%	6.79	2.49	3.31	0.50	7.41	12.74	12.11	2.23	5.29	33.31	10.88	2.88
高水位	极小值	6.90	3.82	0.55	6.97	0.29	3.21	54.21	0.51	38.83	3.40	9.80	76.05
	极大值	7.50	6.21	0.75	8.27	0.33	5.06	55.83	1.08	59.62	10.15	13.8	86.8
	均值	7.10	4.98	0.65	7.56	0.31	4.13	55.18	0.78	48.85	7.50	11.5	81.0
	标准差	0.35	1.19	0.10	0.66	0.02	0.93	0.86	0.29	10.42	3.6	2.07	5.43
	偏度	1.73	0.24	-0.3	0.75	1.48	0.02	-1.45	0.49	0.33	-1.52	1.20	0.69
	变异系数/%	4.88	24.02	15.41	8.70	7.73	22.40	1.55	37.06	21.32	48.01	17.97	6.70

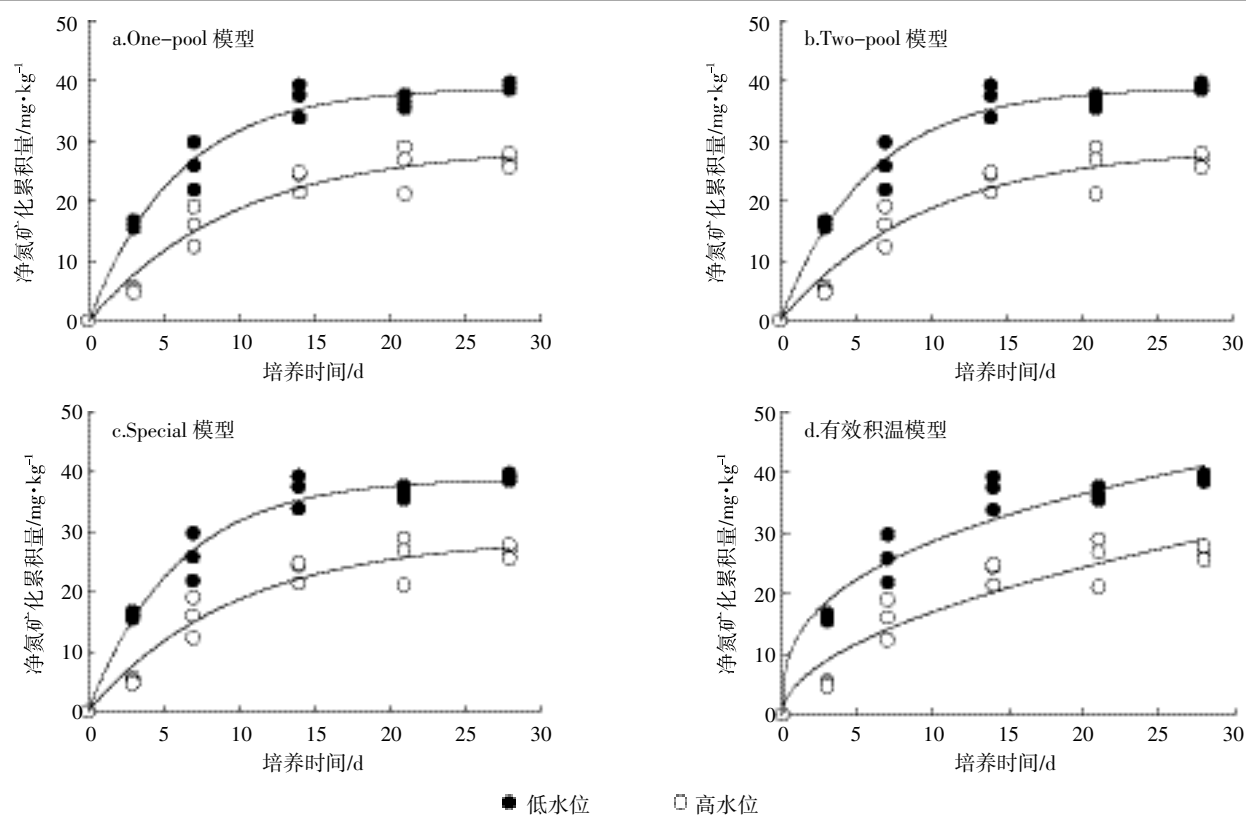


图 3 沉积物氮矿化模型拟合

Figure 3 Model fitting of soil nitrogen mineralization kinetics

表 2 沉积物氮矿化动力学参数拟合

Table 2 Fitted parameters for the nitrogen mineralization kinetic models

水位高程	模型	k	n	$N_d/\text{mg}\cdot\text{kg}^{-1}$	$f_d/\%$	k_d/d^{-1}	R^2	P	RMSE
低水位	One-pool	—	—	38.88	4.05	0.17	0.97	<0.000 1	2.25
	Two-pool	—	—	20.20	2.10	0.17	0.97	<0.000 1	2.43
	Special	—	—	38.55	4.02	0.17	0.96	<0.000 1	2.33
	有效积温	12.79	0.35	—	—	—	0.93	<0.000 1	3.26
高水位	One-pool	—	—	28.98	4.46	0.10	0.93	<0.000 1	2.60
	Two-pool	—	—	14.17	2.18	0.10	0.93	<0.000 1	2.81
	Special	—	—	30.63	4.71	0.12	0.94	<0.000 1	2.56
	有效积温	5.12	0.52	—	—	—	0.88	<0.000 1	3.48

注: f_d 为易矿化氮库矿化势占全氮的比率; 易矿化氮库矿化势 (N_d) = $N \times 10^3 \times f_d$; k_d 为易矿化速率常数; R^2 为模型拟合系数; RMSE 为拟合曲线均方根误差。下同。

Note: f_d is the ratio of N_d to N ; Mineralization potential of easily mineralized nitrogen (N_d) = $N \times 10^3 \times f_d$; k_d is easily mineralization rate constant; R^2 is fitting coefficient of the models; RMSE is the root-mean-square deviation. The same below.

表 3 氮矿化动力学参数与沉积物理化性质相关性

Table 3 Pearson's correlation between kinetic parameters and soil physio-chemical properties

Y	C	N	C/N	P	pH	NH_4^+-N	NO_3^--N	OM	DOC	粘粒	粉粒	砂砾
f_d	-0.995**	-0.932**	-0.998**	0.708	-0.233	-0.952**	0.995**	-0.940**	-0.444	-0.478	-0.956**	0.922**
k_d	0.995**	0.932**	0.998**	-0.708	0.233	0.952**	-0.995**	0.940**	0.444	0.478	0.956**	-0.922**

注: ** 表示在 0.01 水平上显著相关。

Note: ** indicate that correlation is significant at $P < 0.01$ level.

表 4 模型参数预测方程

Table 4 The prediction equation of model parameter

模型参数	预测方程	R_{adj}^2	P	RMSE
f_d	$f_d = 5.09 - 0.07C/N + 0.15OM$	0.985	<0.01	0.006
k_d	$k_d = 0.09 + 0.005C/N - 0.011OM$	0.963	<0.01	0.000 4

3 讨论

澎溪河消落带沉积物 C、N、C/N、 NH_4^+-N 、 $NO_3^- - N$ 含量随水位高程变化差异显著(表 1),其中,C、N、C/N 和 NH_4^+-N 随水位高程降低而增加,而 $NO_3^- - N$ 含量随水位高程降低而降低。可能原因为,一方面,低水位高程淹水胁迫时间较长(年淹水时间为 325 d),缺氧条件下 NH_4^+-N 向 $NO_3^- - N$ 转化受限,且 NH_4^+-N 带正电荷,易被沉积物吸附,不易流失,表现为 NH_4^+-N 累积^[33];另外,沉积物 $NO_3^- - N$ 带负电荷溶水性,使其更易进入水体^[34-35],表现为 $NO_3^- - N$ 流失;同时,还原条件下沉积物碳氮矿化较慢,可能是导致低水位高程 C、N 和 C/N 较高的原因。另一方面,高水位高程淹水时间较短(年淹水时间为 123 d),落干条件下沉积物暴露于空气中, NH_4^+-N 易通过硝化作用转化为 $NO_3^- - N$,且可通过 NH_3 形式挥发而损失^[36-38],表现为 NH_4^+-N 流失、 $NO_3^- - N$ 累积。而植被适生性、多样性和生物量等均随高程增加而增强(大)^[39],可能与沉积物 NH_4^+-N 和 $NO_3^- - N$ 分布存在一定内在联系。另外,沉积物氮矿化累积量培养前期快速上升,后期趋于稳定,这与顾春朝等^[40]所得结果一致。可能原因为,一方面,淹水初期,厌氧微生物迅速繁殖并将有机氮分解为铵态氮。随着培养时间延长,铵态氮积累,厌氧微生物数量饱和。李建兵等^[41]研究表明,过高的铵态氮可能抑制微生物生长,使氮矿化累积量趋于稳定。另一方面,短期培养过程中氮矿化主要来自易分解氮库,这部分氮库受团聚体等物理化学保护较弱,易被优先分解矿化。随着培养时间延长易分解氮消耗殆尽,而沉积物中难分解氮库组分受团聚体等物理化学作用保护较强,难于分解矿化^[42-43]。

四种氮矿化动力学模型均能够较好拟合消落带沉积物氮矿化动力学过程,其中 One-pool 模型对低水位高程拟合 RMSE 值最小,效果最好;Special 模型对高水位高程拟合 RMSE 值最小,效果最好。消落带沉积物氮素矿化过程中,不同水位高程沉积物易矿化氮库矿化势(N_d)存在显著差异(表 2),表现为在低水位高程高于高水位高程,沉积物 C、N、C/N、 NH_4^+-N 、 $NO_3^- - N$ 、OM、粉粒、砂砾与沉积物易矿化氮库矿化势

(f_d)和易矿化速率(k_d)显著相关,受沉积物理化性质影响较大。刘杏认等^[44]研究表明,在一定湿度范围内含水量增加使沉积物氮矿化速率增加。Harrison-Kirk 等^[45]的研究表明土壤质地会影响土壤含水量和通气孔隙,从而影响氮矿化过程。Hanan 等^[13]的研究表明 pH 的变化会对氮矿化过程产生影响。林俊杰等^[46]研究表明,消落带沉积物氨化、硝化及净氮矿化速率与其 N 本底值正相关。氮矿化动力学参数估算表明,C/N 和 OM 是控制模型参数 f_d 和 k_d 的关键因素。Haer 等^[47]研究表明,OM 含量和粘粒比例是影响印度耕地沉积物氮矿化动力学参数估算的主要因素;Schomberg 等^[48]的研究表明 C 和 N 是预测土壤氮矿化潜力的主要因素;周吉利等^[49]研究表明,微生物量碳和 pH 值决定了中亚热带红壤区沉积物的氮矿化过程。此外,本研究尚未考虑季节性温度升高与干湿循环耦合关系对消落带表层沉积物氮矿化动力学过程的影响,在未来的工作中需进一步研究。

4 结论

干湿循环加速了消落带氮矿化动力学过程,增加了低水位高程消落带沉积物易矿化氮重新淹水后大量进入水体的风险,One-pool 模型和 Special 模型分别是低水位和高水位高程氮矿化动力学拟合的最佳模型,其动力学参数与沉积物 C、N、 NH_4^+-N 、 $NO_3^- - N$ 、OM、C/N、粉粒和砂砾显著相关;且 C/N 和 OM 可用于氮矿化动力学模型参数估算,对深入理解三峡支流消落带沉积物氮矿化机制与消落带植被适生性下降、水体富养化之间的关系具有指示意义。

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