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黄河高村至利津河段水体和沉积物中不同形态磷的分布特征

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摘要: 黄河是我国第二大河, 其对营养盐的输送在区域乃至全球范围内扮演着很重要的角色。本研究对黄河高村至利津河段5个站点两季水体和沉积物中不同形态磷的分布特征进行了研究。结果表明, 黄河高村至利津河段水体5月处于磷污染状态, 5月水体总磷(TP)变化范围为 $0.05 \sim 2.31 \text{ mg} \cdot \text{L}^{-1}$, 9月为 $0.03 \sim 0.1 \text{ mg} \cdot \text{L}^{-1}$, 颗粒态磷(PP)为主要赋存形态, TP与PP呈极显著相关($P < 0.01$), 径流量和TP与PP显著正相关($P < 0.05$), 总溶解性磷(TDP)与溶解性无机磷(DIP)显著负相关($P < 0.05, P < 0.01$), 流量增加对TDP和DIP的稀释作用很明显, 但黄河高村至利津河段土壤磷流失仍然很严重。水体悬浮颗粒物(SPM)质量浓度与DIP显著负相关($P < 0.01$), SPM对水体DIP的吸附作用明显; 秋季水体高锰酸盐指数较春季高, 受有机物污染更严重, 水体高锰酸盐指数与DIP质量浓度显著正相关($P < 0.01$)。黄河高村至利津河段沉积物两季总磷的含量基本处于未污染水平, 沉积物TP 5月变化范围为 $284.23 \sim 569.58 \text{ mg} \cdot \text{kg}^{-1}$, 9月为 $287.97 \sim 355.39 \text{ mg} \cdot \text{kg}^{-1}$, 钙磷(Ca-P)为无机磷(IP)的主要赋存形态。活性有机磷(L-OP)含量与径流量, 水体SPM质量浓度和高锰酸盐指数显著相关($P < 0.01, P < 0.05, P < 0.01$), 5月利津站点的有机磷(OP)含量明显较高, 可能是降雨冲刷流域内矿物开采区导致的, 黄河高村至利津河段能源基地生产活动需要密切关注。

关键词: 黄河; 磷; 水体; 沉积物; 磷形态; 富营养化

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Distribution of Different Phosphorus Species in Water and Sediments from Gaocun to Lijin Reaches of the Yellow River

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Abstract: The Yellow River is the second longest river in China, which plays a very important role in the transportation of nutrients at the regional and even global scale. Water and sediment samples were collected at five sites located at the Gaocun to Lijin reaches along the Yellow River and the distribution characteristics of different phosphorus species were analyzed. The concentration of suspended particulate matter (SPM) in water ranged from 1.89 to $2.67 \text{ g} \cdot \text{L}^{-1}$ with an average of $2.26 \text{ g} \cdot \text{L}^{-1}$ in May and from 0.43 to $1.79 \text{ g} \cdot \text{L}^{-1}$ with an average of $1.21 \text{ g} \cdot \text{L}^{-1}$ in September. It almost had the same variation rule as the water flux, which reflects the influence of water flux on SPM transportation. The concentration of total phosphorus (TP) in water ranged from 0.05 to $2.31 \text{ mg} \cdot \text{L}^{-1}$ in May and from 0.03 to $0.1 \text{ mg} \cdot \text{L}^{-1}$ in September. Particulate phosphorus (PP) was the dominant TP species in both months, and TP and PP were significantly correlated ($P < 0.01$), which implied that phosphorus loss in soil from Gaocun to Lijin reaches is still serious. Water runoff was positively correlated with TP and PP ($P < 0.05$), but negatively correlated with total dissolved phosphorus (TDP) and dissolved inorganic phosphorus (DIP) ($P < 0.05, P < 0.01$, respectively). Increasing water runoff obviously increased the dilution of TDP and DIP. The concentration of SPM was negatively correlated with DIP ($P < 0.01$), the adsorption of SPM on phosphorus was mainly the adsorption of DIP. The permanganate index in water was higher in fall suggesting more serious organic pollution and it was positively correlated with DIP ($P < 0.01$). The phosphorus content in sediments was quite low in both months; the total phosphorus (TP) content in sediments ranged from 284.23 to $569.58 \text{ mg} \cdot \text{kg}^{-1}$ in May and from 287.97 to $355.39 \text{ mg} \cdot \text{kg}^{-1}$ in September. Calcium-bound phosphate (Ca-P) was the dominant species of inorganic phosphorus (IP). The labile organic phosphorus (L-OP) content was significantly correlated with runoff, SPM concentration, and permanganate index ($P < 0.01, P < 0.05, P < 0.01$, respectively). The organic phosphorus (OP) content was significantly higher at the Lijin site in May, likely caused by the rainfall-runoff from the mining area around Lijin. The energy-based production activities from the Gaocun to Lijin reaches should also be further investigated.

Key words: Yellow River; phosphorus; water; sediments; phosphorus fraction; water eutrophication

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在过去的几十年中,随着世界人口的快速增长,农业生产与能源消耗很大程度上改变了土地利用方式,对全球淡水资源产生了不良影响. 人类活动产生的相当一部分营养因子进入水体中,被河流输送进入海洋. 河流的营养盐输送有利于维持海洋初级生产力,但过剩则会引起富营养化污染现象^[1]. 对近海而言,河流磷输入是其最重要的磷输入来源,河流向近海的磷输送不可忽视^[2]. 河流中的营养盐含量不仅随径流量的变化存在年际和年内的差别,也与沿岸土地利用方式和农业施肥密切相关,密西西比河口淡水段 DIN 与 $\text{PO}_4^{3-}\text{-P}$ 的比值具有冬春季较高,夏秋季较低的特点^[3]. 西班牙 Ebro 河每年向地中海输送 200 t 磷酸盐^[4],英国 Tweed 河口淡水端 $\text{PO}_4^{3-}\text{-P}$ 的季节起伏比较大^[5],中国长江无机磷的入海浓度有升高趋势^[6],大辽河淡水无机磷质量浓度在平水期很高^[7]. 有研究表明从 1980 年代初至 1990 年代末渤海的生态系统的营养盐结构发生了很大变化,氮磷比升高,渤海的磷很大一部分由黄河输入^[8]. 而近期黄河下游研究中一些数据表明数十年来黄河下游的磷含量较高^[9~11].

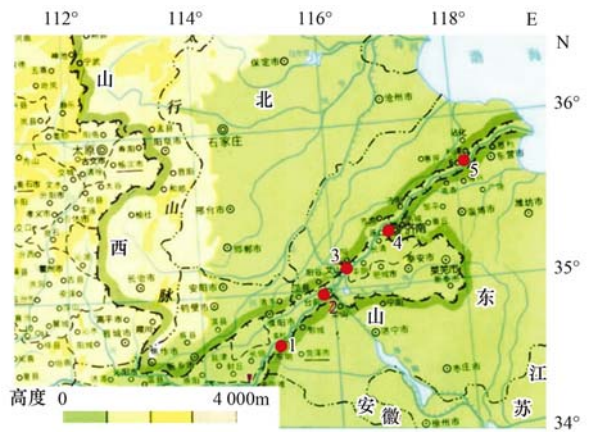
本研究从多形态磷的角度出发,对两季黄河水体和沉积物进行磷分布特征的对比并分析其主要来源,有助于实现黄河水质好转和水资源的合理利用,以为黄河下游和入海流域污染总量控制提供重要依据.

1 材料与方法

1.1 样品采集

按照国家环境保护总局“地表水和污水检测技术规范(HJ/T 91-2002)”,于 2015 年 5 月和 9 月,在黄河下游自高村站点至河口段山东东营利津站点采集实验所需的水样和沉积物样品. 具体采样点位如图 1 所示,用便携式采水器在水面以下约 50 cm 处采集黄河水体水样,将水样缓慢注入 500 mL 聚乙烯样品瓶,当溢出水样体积超过样品瓶体积约 2 倍时加盖并迅速运回实验室.

黄河沉积物样品采集与水体样品采集同步进行. 用有机玻璃圆柱状采样柱($d=5\text{ cm}, h=40\text{ cm}$)采集采样点表层未扰动沉积物,采样厚度不小于 20 cm,每个点位采集 4 根平行样. 采样柱上端保留上覆水,两端用橡皮塞塞紧后垂直安置. 带回实验室后,去除采样柱上端上覆水,将 4 根平行样的采样柱表层 10 cm 沉积物混合均匀,并剔除其中的动植物残体,经风干研磨过筛后用于测定各形态磷含量.



1. 高村; 2. 孙口; 3. 艾山; 4. 涿口; 5. 利津;
比例尺为 1:6 000 000

图 1 黄河高村至利津河段采样站点分布示意
Fig. 1 Sampling sites from the Gaocun to Lijin reaches along the Yellow River

1.2 样品及数据分析

1.2.1 水样分析

采样断面流量用超声波流量计(F601/G601)检测,水温用便携式溶氧仪(JPB-608)检测,pH 用便携式 pH 计(FG2-FK)检测,高锰酸盐指数用碱性高锰酸钾法测定. 悬浮颗粒物浓度(SPM)由重量法测定,总磷(TP)、总溶解性磷(TDP)、溶解性无机磷(DIP),颗粒态磷(PP)的测定方法见表 1.

表 1 水样分析项目

Table 1 Measurements for water samples

项目	测定方法
TP	钼酸铵分光光度法 GB 11893-89
TDP	钼酸铵分光光度法 GB 11893-89 样品过膜(0.45 μm)消解
DIP	钼酸铵分光光度法 GB 11893-89 样品过膜(0.45 μm)不消解
PP	TP-TDP

1.2.2 沉积物分析

采用化学连续提取法测定沉积物样品中各形态无机和有机磷含量^[12,13]. 无机磷(IP)分为 5 种形态:交换态磷(Ex-P)、铝磷(Al-P)、铁磷(Fe-P)、闭蓄态磷(Oc-P)和钙磷(Ca-P); 有机磷(OP)分成 4 种形态:活性有机磷(L-OP)、中活性有机磷(ML-OP)、中稳定性有机磷(MR-OP)和高稳定性有机磷(HR-OP).

1.2.3 数据分析

数据通过 Excel 2010 和 Origin 9.0 进行数据整理和图形绘制,用 SPSS 20 进行统计分析.

2 结果与讨论

2.1 水体基本理化性质

采样河段各采样点 5 月和 9 月水体 pH 基本没

有变化,在 8.1~8.2 之间浮动,属于碱性水. 5 月水温变化范围为 15.1~17.1℃,平均为 16.1℃; 9 月水温变化范围为 15.7~19.8℃,平均为 16.7℃,两季水体温度差异也较小. 水体 SPM 的质量浓度不高,5 月为 1.89~2.67 g·L⁻¹,平均值为 2.26 g·L⁻¹. 9 月为 0.43~1.79 g·L⁻¹,平均值为 1.21 g·L⁻¹. 5 月黄河高村至利津段径流量远远大于 9 月 [图 2(a)],经过涑口之后,两季利津站点 SPM 和径流量均明显减少. 由 2015 年中国河流泥沙公报可知,径流量减少造成黄河下游涑口-利津段河段淤积,水体流速减慢,悬浮颗粒物沉降效率增加,因而 SPM 质量浓度明显降低. SPM 分布与径流量的变化较为一致,反映了水流对悬浮物输送的影响^[14]. 两季水体中高锰酸盐指数的空间分布变化不大,5 月为 2.20~2.40 g·L⁻¹,平均值为 2.30 g·L⁻¹. 9 月为 3.10~3.50 g·L⁻¹,平均值为 3.30 g·L⁻¹,秋季水体中高锰酸盐指数质量浓度明显升高 [图 2(b)],说明黄河高村至利津河段水体秋季较春季受有机物污染严重.

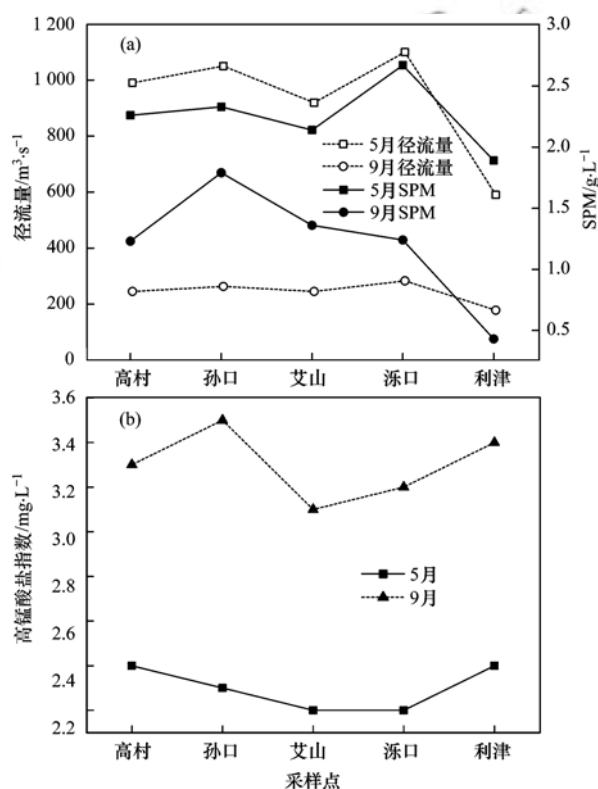


图2 黄河高村至利津河段 SPM、流量和高锰酸盐指数的沿程分布

Fig. 2 Distribution of SPM, flux, and permanganate index from the Gaocun to Lijin reaches along the Yellow River

2.2 水体中不同形态磷的分布特征

水体总磷及各形态磷的质量浓度见图 3. 5 月

水体 TP 变化范围为 0.05~2.31 mg·L⁻¹, 9 月为 0.03~0.1 mg·L⁻¹. 根据国家《地表水环境质量标准》(GB 3838-2002), 5 月采样河段除艾山点外均为劣 V 类水体, 9 月均为 II 类以上水体 (以 TP 计), TP 质量浓度与张晓晓等的研究相似^[15]. 水体中 TP 5 月和 9 月均以 PP 为主要赋存形态, TP 与 PP 浓度在 $P < 0.01$ 水平呈极显著相关 (表 2), 由图 3(a) 可知, 艾山站点的 PP 含量明显低于其它站点, 导致 TP 含量也明显偏低. 艾山采样断面位于黄河干流和大汶河交汇处的下游, 支流汇入处流速降低, 水体中一部分颗粒物沉降 (从图 2 可知该站点 SPM 降低), 因此 PP 含量明显低. 5 月各采样点水体 PP 占 TP 的 86%~99%, TDP 浓度较低 (平均 0.01 mg·L⁻¹). 9 月除艾山站点外, PP 对 TP 贡献率仅为 62%~84% [图 3(a)], 可能径流量的降低导致河道流速减慢, 使悬浮颗粒物沉降 (图 2), 从而导致 PP 减少而 TDP 含量增加 (平均 0.02 mg·L⁻¹). 由表 2 可知两季 TDP 与 DIP 质量浓度极显著相关 ($P < 0.01$), DIP 主要由农业施肥、城市污水和大气的干湿沉降引起, 且磷酸根易结合形成难溶的化合物, 不易流失^[16]; 水体 DOP 质量浓度 5 月和 9 月变化不大 [图 3(b)].

对水体理化性质与水体各形态磷进行的相关性分析表明, pH 和水温对水体各形态磷质量浓度影响不大. 而径流量和 TP 和 PP 显著正相关 ($P < 0.05$), 和 TDP 与 DIP 分别在 $P < 0.05$ 和 $P < 0.01$ 的条件下显著负相关, 流量增加对 TDP 和 DIP 的稀释作用很明显, PP 与 SPM、径流量三者的变化规律相似, 说明黄河该河段土壤磷的流失仍是 PP 含量高的一大原因^[17]. SPM 质量浓度与 DIP 显著负相关 ($P < 0.01$), 可见悬浮颗粒物对水体磷的吸附主要是对 DIP 的吸附, 水体 SPM 含量降低, DIP 的吸附量减少, 因此水体 DIP 含量增加, 符合两季水体 SPM 和 DIP 的变化规律. 水体高锰酸盐指数与 TDP 尤其 DIP 显著正相关 ($P < 0.05$, $P < 0.01$), 这是因为高锰酸盐指数和 DIP 通常具有同源性, 陆地径流输入和陆源排放是其主要来源^[18], 因此高锰酸盐指数在一定程度上可以表征陆源排入河流带来的磷含量 (图 4).

2.3 沉积物中不同形态磷的分布特征

沉积物中 TP 5 月含量变化范围为 284.23~569.58 mg·kg⁻¹, 9 月为 287.97~355.39 mg·kg⁻¹, 根据刘鸿亮等的研究^[19], 将沉积物分为未污染 [$\omega(\text{TP}) \leq 500 \text{ mg} \cdot \text{kg}^{-1}$], 中度污染 [$500 \text{ mg} \cdot \text{kg}^{-1}$

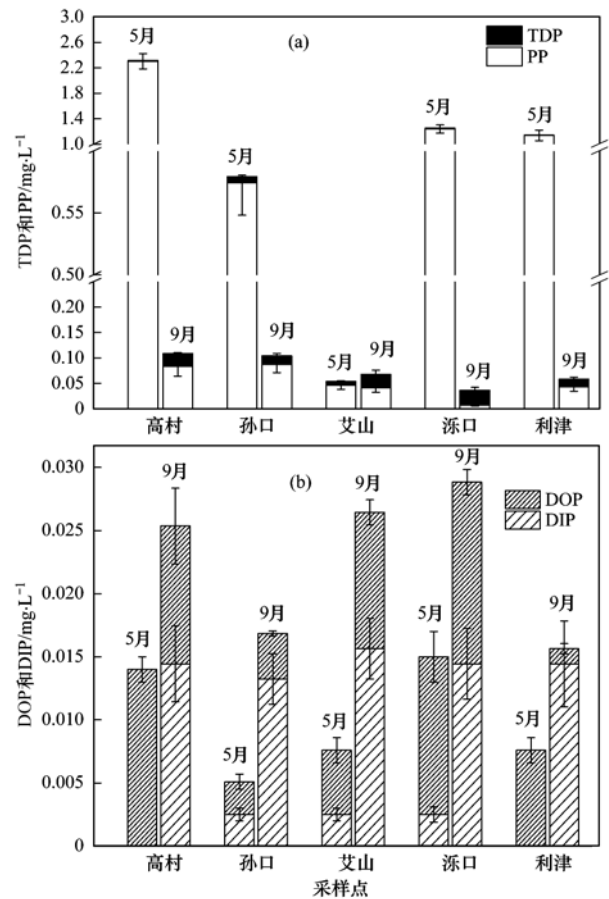


图3 黄河高村至利津河段水体各形态磷浓度分布
Fig. 3 Concentration of phosphorus fractions from Gaocun to Lijin reaches along the Yellow River

表2 黄河高村至利津河段水体各形态磷相关性分析结果¹⁾

Table 2 Correlation analysis of different phosphorus species in water from Gaocun to Lijin reaches along the Yellow River

	TP	PP	TDP	DIP
TP	1			
PP	0.99 **	1		
TDP	-0.37	-0.37	1	
DIP	-0.75 *	-0.76 *	0.80 **	1

1) **表示 $P < 0.01$ 时显著相关, * 表示 $P < 0.05$ 时显著相关, $n = 10$

$\leq \omega(\text{TP}) \leq 1\,000\text{ mg}\cdot\text{kg}^{-1}$] 和严重污染 [$\omega(\text{TP}) > 1\,000\text{ mg}\cdot\text{kg}^{-1}$]. 黄河该河段各点沉积物 5 月除利津属于中度磷污染状态,其他站点均为未污染状态. 9 月所有站点沉积物均为未污染状态. 由表 3 可知,各形态磷与 TP 相关显著性系数由大到小分别为 $\text{Ca-P} > \text{ML-OP} > \text{HR-OP} > \text{Al-P} > \text{Fe-P} > \text{Ex-P} > \text{Oc-P} > \text{L-OP} > \text{MR-OP}$,排在前 3 位的 Ca-P、HR-OP 分别是最稳定的无机磷和有机磷形态,而 ML-OP 活性也不高,不易转化为无机磷被生物吸收. 说明黄河该河段沉积物对上覆水体的磷污染潜力不高.

5 月和 9 月,IP 均为 TP 的主要赋存形态,分别平均占 TP 的 90% 和 96%. 黄河下游沉积物各形态无机磷含量分布见图 5(a). Ca-P 则为 IP 的主要赋存形态,占 IP 的 96% 以上,Ca-P 是沉积物中比较稳定的磷形态,常深埋于沉积物中而难于被藻类等浮

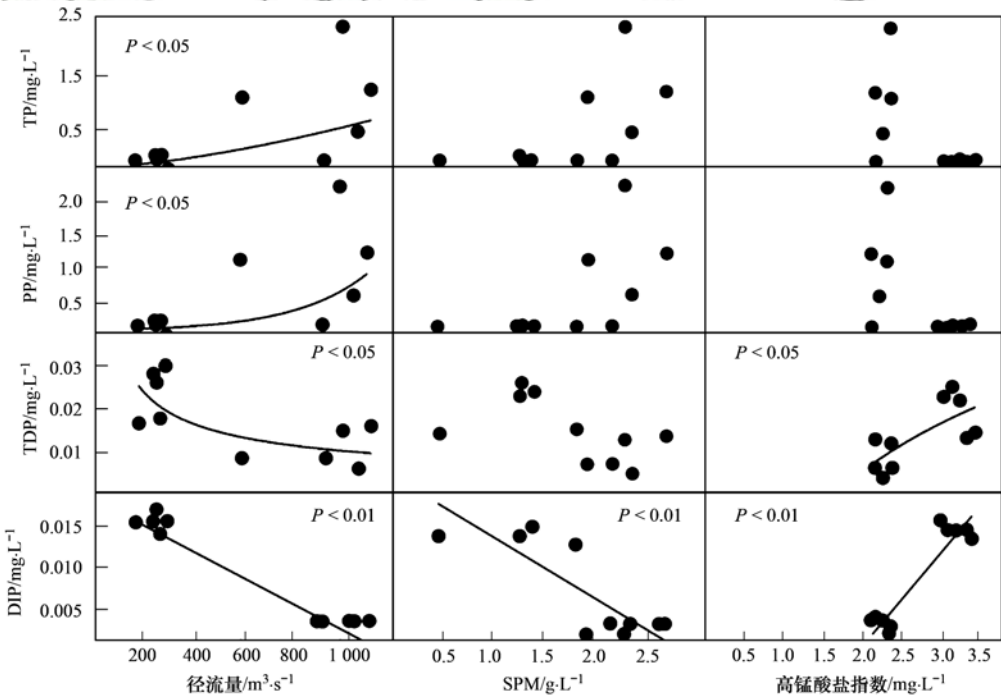


图4 水体理化性质与水体各形态磷相关性分析

Fig. 4 Correlation analysis of different phosphorus species in water and physicochemical properties of water

表 3 黄河高村至利津河段沉积物中各形态磷相关性分析结果¹⁾

Table 3 Correlation analysis of phosphorus fractions in sediments from Gaocun to Lijin reaches along the Yellow River

	TP	Ex-P	Al-P	Fe-P	Oc-P	Ca-P	L-OP	ML-OP	MR-OP	HR-OP
TP	1									
Ex-P	0.29	1								
Al-P	0.45	0.56	1							
Fe-P	0.40	0.55	0.69 *	1						
Oc-P	0.06	0.06	0.08	0.69 *	1					
Ca-P	0.89 **	0.36	0.53	0.56	0.17	1				
L-OP	-0.05	0.08	-0.14	0.45	0.85 **	0.09	1			
ML-OP	0.82 **	0.05	0.22	0.03	-0.17	0.50	-0.31	1		
MR-OP	-0.25	-0.75 *	-0.57	-0.74 *	-0.27	-0.28	-0.02	-0.13	1	
HR-OP	0.80 **	0.23	0.23	0.09	-0.12	0.46	-0.22	0.90 **	-0.017	1

1) ** 表示 $P < 0.01$ 时显著相关, * 表示 $P < 0.05$ 时显著相关, $n = 10$

游生物所利用,一般含量较高^[20,21]. 被无定形铁、铝、钙等胶膜所包蔽的 Oc-P 除在 5 月艾山和涑口含量较高以外,其他点均含量较低,可能是由于这两点同时位于大汶河流域铁、煤、铝矿开采区,5 月径流量大,矿业污染通过冲刷大量进入河流造成的. 两季吸附在铁、铝氧化物和氢氧化物胶体表面上的 Fe-P、Al-P 以及易被生物利用的 Ex-P 含量相近,含量均 $< 2.00 \text{ mg} \cdot \text{kg}^{-1}$. Fe-P 与 Al-P 和 Oc-P 之间分布均存在显著正相关性 ($P < 0.05$),可能在各种生物地球化学作用下,不同胶体表面由于吸附解吸效率的不同导致了不同形态磷含量的变化.

OP 包括由陆源性排放物质组成的难降解性有机磷和由死亡的水生生物尸体组成的可降解性有机磷,与人类活动有关,主要来源于面源污染^[22]. 黄河下游沉积物各形态 OP 含量分布见图 5(b). 除 5 月利津点外,其他站点两季春秋 OP 含量不高,变化范围为 $5.88 \sim 29.40 \text{ mg} \cdot \text{kg}^{-1}$. 5 月利津采样断面 ML-OP 和 HR-OP 是有机磷的主要赋存形态,分别是其他采样点的 10 倍和 4 倍以上. ML-OP 是活性仅次于活性有机磷的有机磷形态,来源于间隙水或者以物理吸附态附着于碳酸盐、氧化物、氢氧化物或存在于黏土矿粒等其他相上,HR-OP 作为稳定性较高的有机磷形态,其主要成分包括腐殖质中胡敏酸和胡敏素中的有机磷、肌醇含 4~6 个磷酸根的铁铝盐或者少量被 Fe_2O_3 胶膜所包裹的铝磷或钙磷^[23]. 5 月利津采样断面这两种形态有机磷含量高可能是由外源 P 污染输入造成的^[24],这进一步印证了 5 月大汶河流域矿物开采造成沉积物磷污染的推断,可见控制汛期黄河入海口利津周边的能源基地生产活动是控制沉积物磷污染的必要措施.

对水体理化性质与沉积物各形态磷含量进行相关性分析,发现水温, pH 对沉积物各形态磷含量变

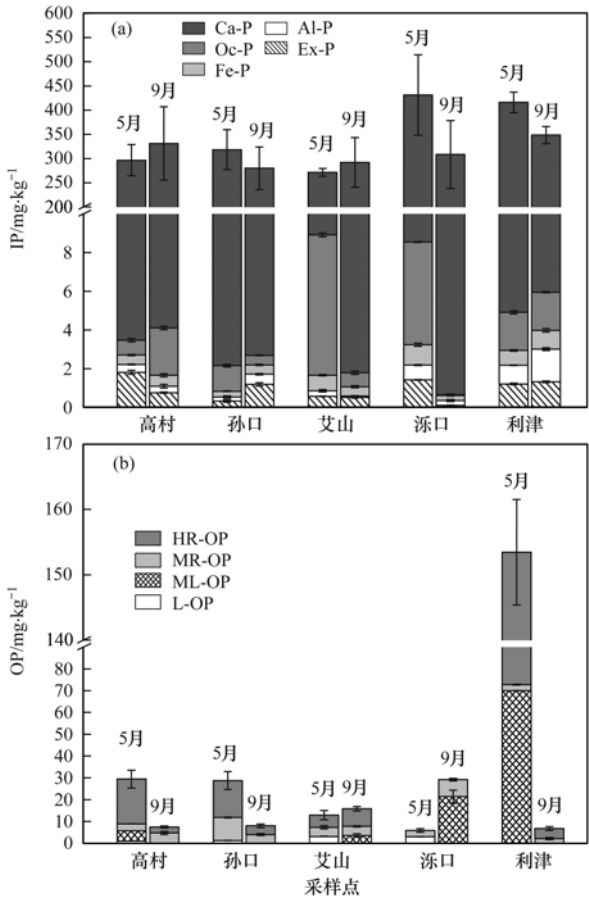


图 5 黄河高村至利津河段沉积物中各形态无机磷和有机磷含量分布

Fig. 5 Inorganic and organic phosphorus fractions in surface sediments from Gaocun to Lijin reaches along the Yellow River

化没有很大影响,仅 L-OP 与水体理化性质有相关关系(图 6). 由图 6 可知,水体径流量、SPM 质量浓度与沉积物 L-OP 含量均正相关 ($P < 0.01$, $P < 0.05$),说明径流量和水体 SPM 的增加携带的有机磷污染除了影响水体磷含量变化,也会造成沉积物有机磷,尤其是 L-OP 的增加. L-OP 是沉积物中最

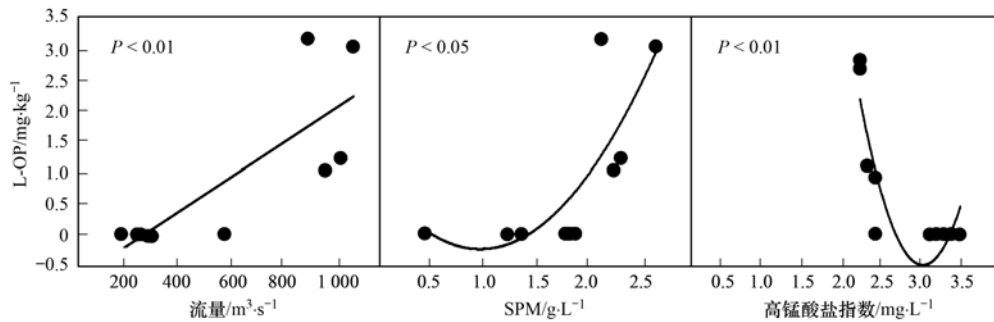


图6 水体理化性质与沉积物 L-OP 含量的相关性分析

Fig. 6 Correlation analysis of L-OP in sediments and physicochemical properties of water

具有释放潜力的有机磷,矿化分解很快,极易成为上覆水中藻类生长的磷源^[25],水体高锰酸盐指数与沉积物 L-OP 含量呈负显著相关($P < 0.01$),可见水体中有机污染物浓度增加会一定程度上刺激沉积物作为磷源向水体释放磷,造成水体溶解态磷增加(图4),以达到适宜的营养比例。

3 结论

(1)5月黄河高村至利津河段径流量远远大于9月,两季利津点径流量的减少造成冻口-利津河段淤积,入海口利津 SPM 质量浓度显著降低,秋季水体中高锰酸盐指数质量浓度明显升高,较春季受有机物污染严重。

(2)5月黄河高村至利津河段水体磷污染严重。5月和9月 PP 均为 TP 的主要赋存形态,TP 与 PP 质量浓度在 $P < 0.01$ 水平呈极显著相关,PP 与 SPM 和径流量三者的变化规律相似,表明黄河该河段土壤磷流失仍然严重。水体 DIP 与径流量和 SPM 质量浓度均呈极显著负相关关系($P < 0.01$),与高锰酸盐指数呈极显著正相关($P < 0.01$),说明径流量对 DIP 的稀释及 SPM 对 DIP 的吸附作用明显,高锰酸盐指数和 DIP 可能具有同源性。黄河该河段面源污染和陆源排放的控制十分必要。

(3)黄河高村至利津河段沉积物磷含量基本处于未污染状态,两季 Ca-P 均为 IP 的主要赋存形态,径流量,水体 SPM 质量浓度和高锰酸盐指数对沉积物中 L-OP 含量有一定影响。5月利津采样断面 ML-OP 和 HR-OP 含量明显较高,可能是降雨冲刷流域内矿物开采区导致的。

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