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运用硫同位素、氮氧同位素示踪里湖地下河硫酸盐、 硝酸盐来源

李瑞1,2,肖琼2*,刘文1,2,4,郭芳2,潘谋成2,于奭2,3

(1. 西南大学地理科学学院,重庆 400715;2. 中国地质科学院岩溶地质研究所,联合国教科文组织国际岩溶研究中心,国土资源部/广西岩溶动力学重点实验室,桂林 541004;3. 中国地质大学环境学院,武汉 430074;4. 山东省地矿工程勘察院,济南 250014)

摘要:为揭示里湖地下水水质的时空变化规律,结合硫同位素及氮氧同位素分析水质变化原因,为开发和保护岩溶水资源提供科学依据.于 2010年1月至12月,2014年5月、10月对里湖地下河进行监测,分析其常规水化学组成及 δ^{34} S-SO $_4^2$ 、 δ^{15} N-NO $_3$ 、 δ^{18} O-NO $_3$ 特征.结果表明:①地下河水化学类型为 HCO $_3$ -Ca 型,受到大气降水、人类活动等影响,各离子表现出明显的时空变化规律;②自污水处理厂运营以来,地下河中硫酸盐浓度略有降低,但仍然偏高,与 2010年相比,硝酸盐污染加剧.受人类活动影响上游拉易洞、南丹河,中游凉风洞、甘田坝等地硫酸盐、硝酸盐浓度较高,下游小龙洞处浓度较低;③地下河 δ^{34} S-SO $_4^2$ 值在 -4.12% \sim -0.93% 之间,通过与潜在硫源的 δ^{34} S比对,推断由工业和居民生活燃煤产生的大量硫氧化物引起雨水酸化,以酸雨的形式向当地地下水输入大量 SO $_4^2$;④ δ^{15} N-NO $_3$ 值在 0.26% \sim 11.58% 之间,平均为 7.61% $,\delta^{18}$ O-NO $_3$ 在 -2.33% \sim 21.76% 之间,平均为 9.38% ,结合硝酸盐氮氧同位素组成分析,认为土壤氮、人畜粪便和污水是研究区硝酸盐的主要贡献者,也是地下水中硝酸盐污染加剧的主要原因.

关键词:岩溶地下河;水化学;硫同位素;氮氧同位素;示踪来源

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Using $\delta^{34}S$ -SO₄²⁻ and $\delta^{15}N$ -NO₃⁻, $\delta^{18}O$ -NO₃⁻ to Trace the Sources of Sulfur and Nitrate in Lihu Lake Undergound Water, Guangxi, China

LI Rui^{1,2}, XIAO Qiong^{2*}, LIU Wen^{1,2,4}, GUO Fang², PAN Mou-cheng², YU Shi^{2,3}

(1. School of Geographical Sciences, Southwest University, Chongqing 400715, China; 2. Key Laboratory of Karst Dynamics, Ministry of Land and Resources/ Guangxi, International Research Center on Karst Under the Auspices of UNESCO, Institute of Karst Geology, Chinese Academy of Geological Sciences, Guilin 541004, China; 3. School of Environmental Studies, China University of Geosciences, Wuhan 430074, China; 4. Shandong Geological Engineering Investigation Institute, Ji'nan 250014, China)

Abstract: To reveal the temporal and spatial variation pattern of groundwater chemistry in Lihu Lake and explore the causes for the change of water quality through analysis of sulfur isotope and nitrogen-oxygen isotope, so as to provide scientific basis for reasonable exploitation and protection of karst water resources. Several groundwater samples, collected from January to December in 2010 and May, October in 2014 were monitored to analyze the chemical composition of conventional water and the characteristics of $\delta^{34}S-SO_4^{2-}$, $\delta^{15}N-NO_3^{-}$, and $\delta^{18}O-NO_3^{-}$. The results showed that: ① The hydrochemical type of the underground water was HCO_3 -Ca type and effected by the seasonal precipitation and human activity, the temporal and spatial variation of the main cations was obvious. ② The sulfur concentration in the underground river was slightly decreased since the operation of the sewage plant, however, the sulfur concentration was still high. The nitrate pollution aggravated in the year 2014 compared to 2010. Impacted by human activity, the concentration of sulfur and nitrate was higher in the upstream Nandan river, Layi cave and the midstream Liangfeng cave, and Gantianba than in the downstream Xiaolong cave. ③ The $\delta^{34}S-SO_4^{2-}$ value ranged from -4. 12 ‰ to -0. 93 ‰. It was inferred that the emission of sulfur oxides through burning coal resulted in the rainwater acidification, which input a large amount of SO_4^{2-} into the underground water in the form of acid rain. ④ The $\delta^{15}N-NO_3^{-}$ value ranged from 0. 26 ‰ to 11. 58 ‰, with an average value of 7. 61 ‰, the $\delta^{18}O-NO_3^{-}$ value ranged from -2. 33 ‰ to 21. 76 ‰, with an average value of 9. 38 ‰. In combination of the composition analysis of nitrogen-oxygen isotope of nitrate, it was believed that soil organic nitrogen, manure and sewage were the main sources of nitrate in the groundwater and the main causes for aggravation of nitrate pollution of the underground water.

Key words: karst underground water; hydrochemistry; sulfur isotope; nitrate-oxygen isotope; trace source

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作者简介: 李瑞(1990~),女,硕士研究生,主要研究方向为岩溶水文地质学,E-mail;616569272@qq.com

^{*} 通讯联系人, E-mail: xiaoqiong-8423@163. com

人类活动向地下河中排放的污染物质往往具有 多样性和多源性,运用单一的指标判别地下水中所 有污染源往往缺乏针对性和科学性,因此,需要结合 多种同位素分别示踪污染物质来源. 地下水中硝酸 盐污染的来源较多,早期主要利用氮同位素技术识 别氮源^[1~6],但由于不同污染源 NO; 的初始δ¹⁵N值 存在交迭现象,以及诸如反硝化作用等地球化学作 用引起同位素分馏,利用单一的δ¹⁵N值难以区 $\mathcal{G}^{[7,8]}$. 为此,将 δ^{15} N-NO, 和 δ^{18} O-NO, 相结合能有 效地确定污染来源,识别反硝化作用[9~16]. 而针对 地下河中硫酸盐来源问题,根据代表不同来源污染 物特征化学指纹的δ³⁴S值,在没有硫同位素分馏效应 以及已知潜在硫源的δ³⁴S的情况下,可以通过所测样 品的δ³⁴S推测其硫源,较好地进行示踪^[17~21]. 基于 此,本研究在监测里湖地下河各常规离子时间变化 特征的基础上,联系城市发展情况和流域的水文地 质条件,利用硫同位素示踪硫酸盐来源,氮氧同位素 示踪硝酸盐来源,以期为厘清城市污染物质来源,从 源头上减少硫酸盐、硝酸盐输入,协调城市经济发 展与水质保护提供科学依据.

1 研究区概况

里湖地下河位于广西河池市南丹县中北部,是 区内最大的地下河水系,其流域面积为517.4 km², 其中碳酸盐岩出露面积占总面积的85%左右(图 1). 里湖地下河发源于拉易、关西的峰林、峰丛溶 盆-溶洼地区,水系呈树枝状展布,径流于峰林溶洼 山地,于贵州省荔波县拉娘村西北打狗河西岸陡崖 脚出露排泄. 流域内出露地层主要为石炭系、二叠 系、三叠系,仅在南丹河河谷地带存在第四系堆积 物,广泛分布含燧石石灰岩,偶夹煤层及白云岩,灰 岩,砂岩,页岩等. 研究区为中亚热带季风性湿润气 候,年平均气温为 17~20℃,月平均最高气温为 27℃,最低气温7℃.本区降水充沛,据南丹县1965 ~2009年以来45年的降水数据显示,降水主要集 中在 5~8 月, 达 965.2 mm, 占全年降水量的 65.7% [22], 是地下水的主要补给来源之一[23]. 受降 水季节变化的影响,地下河平水期流量4593.75 L·s⁻¹, 枯季流量3 339.66 L·s⁻¹, 其中地表水补给流 量1 259. 05 L·s⁻¹.

里湖地下河是南丹县兼有城市供水、农业灌溉,工业用水等多重社会和经济功能的重要水源(图1).由于90年代以来城市化进程的加快,城市工业废水,生活污水和人畜废水大量排放,里湖地下

河污染载荷增加,水质变差. 已有研究表明氨氮是里湖地下河主要污染指标,亚硝态氮、锰、总铁、化学需氧量(chemical oxygen demand, COD)、生化需氧量(biochemical oxygen demand, BOD)部分浓度超过国家II类水标准,砷、铬、硝态氮的部分浓度值超过国家II类水质量标准,部分采样点水污染严重,已基本不能利用[23]. 郭芳等[24]选取水中主要离子和微量元素,分析了里湖地下河的自净能力,为地下河水资源管理和保护提供了依据. 上述研究从不同角度对里湖地下河的水质进行了分析,指出人类活动是里湖地下河水质变化的因素,但得出的污染源的结果较为粗略,要从源头上减少污染物质的输入,还需要利用同位素示踪方法直接、明确地追踪出污染来源.

2 材料与方法

在里湖地下河共布设7个采样点(图1),采样工作分3个时间段,即2010年1~12月(马振杰^[23]的采样)、2014年5月16日、2014年10月18日. 其中,在小场采集里湖地下河上游的南丹河水样,该地表水流经南丹县城,途经居民区,菜市场等,生活和生产废水被直接或间接地排放到南丹河,对南丹河的水质产生直接影响. 拉易洞为其上游地表河转化为地下河的结点,是里湖地下河的主要人口之一. 凉风洞、甘田坝、红星河等是里湖地下水的出露点,其间明暗流交替. 桥村是南丹县的备用水源地,根据柳州水文队调查,认为该股水与拉易洞水为上下层关系,最终将汇入里湖地下河. 小龙洞是里湖地下河汇入打狗河的出口.

现场用 WTW 3430 多参数水质测定仪(德国 WTW 公司)测定水温、溶解氧(dissolved oxygen, DO)、pH 值, 电导率, 精度分别为 0.1 °C、0.01 mg·L⁻¹、0.01 个 pH 单位、1 μ S·cm⁻¹; HCO₃ 和 Ca²+采用德国 Merck 公司便携式试剂盒, 精度分别为 0.1 mmol·L⁻¹和 2 mg·L⁻¹.

每次采样前,用水样清洗采样瓶 3~4 次,现场用直径 50 mm、0.45 μm 的醋酸纤维滤膜过滤水样后,存储于 100 mL 聚乙烯采样瓶中,带回实验室用离子色谱分析仪测定其 SO_4^{2-} 、 NO_3^{-} 、 Cl^{-} ,此测试工作在广西/国土资源部岩溶动力学重点实验室完成;取过滤水样,装于事先用水样清洗过的 50 mL 聚乙烯样瓶中,并立即加 1:1优级纯硝酸溶液 5~8 滴,调节 pH < 2,用美国 Perkin Elmer 公司产的 Optima 2100DV 全谱直读型 ICP-OES 测定水样中的 K⁺、

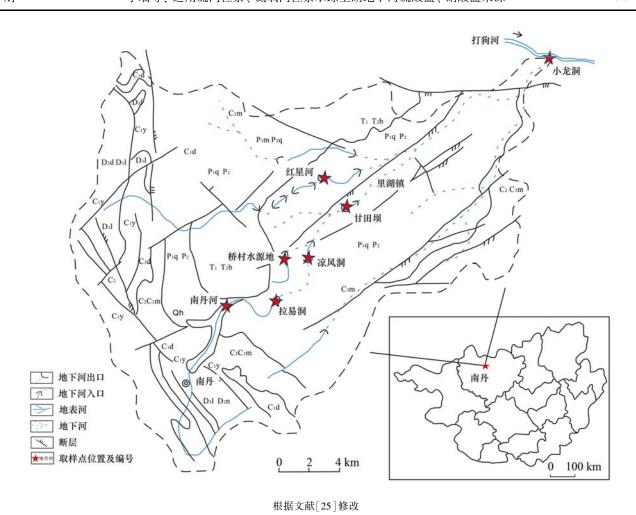


Fig. 1 Hydrogeological map of the studied area and the locations of sampling sites

图 1 研究区水文地质图及采样点位置

Na⁺、Mg²⁺,阳离子测试在西南大学地理科学学院 地球化学同位素实验室完成.

水中 SO_4^{2-} 的 δ^{34} S在 2014 年 5 月 16 日采集,现场用 0.45 μ m 醋酸纤维滤膜过滤 1.5 L 水样装入事先清洗 3 遍的采样瓶. 带回实验室后,加HCl 酸化,使 $pH \leq 2$,视水样中 SO_4^{2-} 浓度加入过量的饱和 $BaCl_2$ 溶液,使 SO_4^{2-} 完全生成 $BaSO_4$,静置过夜. 用去离子水反复冲洗沉淀保证完全去除 Cl^- ,并用 0.45 μ m 醋酸纤维滤膜过滤生成的 $BaSO_4$ 沉淀,置于 850 C 烘干 2 h. δ^{34} S- SO_4^{2-} 采用元素分析仪(Carlo Erba 1108)结合同位素质谱仪(IRMS,Delta C Finningan Mat)测定,测定结果相对于国际标准 V-CDT,测试精度优于 ± 0.2 ‰,测试工作在中国地质大学(武汉)环境学院教学实验中心完成.

用事先清洗 3 遍的 50 mL 聚乙烯瓶取水样,密封,冷藏送实验室进行水中δ¹5N-NO₃ 和δ¹8O-NO₃ 同位素分析,样品用反硝化细菌法结合痕量气体分析

仪(TraceGas)/同位素比质谱仪进行测试.利用天然存在无 N_2O 还原性酶活性的反硝化细菌将 NO_3 转化为 N_2O 气体,再采用痕量气体分析仪 TraceGas在线提取和纯化 N_2O 气体,利用含高氯酸镁及碱石棉的水阱除去水分和 CO_2 ,再用液氮捕集浓缩 N_2O 气体,然后加热释放,经 Porapak Q 色谱柱分离后由He 气带入同位素比质谱仪进行测定,测试工作在中国农业科学院农业环境与可持续发展研究所稳定同位素实验室完成.

3 结果与分析

3.1 水化学类型

表 1 为里湖地下河部分测试数据,图 2 为研究区 7 个采样点的水化学 Piper 三线图,在阳离子三角图上,里湖地下河阳离子以 Ca^{2+} 为主,平均占阳离子组成的 74%,阴离子三角图上以 HCO_3 为主要离子,平均占阴离子组成的 71%,其次为 SO_4^{2-} ,平均占阴离子组成的 24%,水化学类型为 HCO_3 -Ca 型. 从

阳离浓度均值来看, Ca²⁺ > Na⁺ > Mg²⁺ > K⁺; 从阴 离子浓度均值来看,HCO; >SO₄ > NO; >Cl .

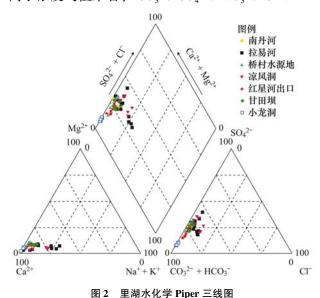


Fig. 2 Piper chart of Lihu Lake underground water

3.2 里湖水化学时间变化特征

里湖地下河电导率变化在 231~803 μS·cm⁻¹, pH 在 7. 10~8. 87 之间,水温变化在 13. 5~24. 7℃ 之间,受气温影响,夏季大于冬季. 分别选取代表里 湖地下河上、中、下游的拉易洞、凉风洞、小龙洞 来分析里湖地下河水化学时间变化(图3). 上游拉 易洞和中游凉风洞各离子浓度时间变化上基本相 同,即 K⁺、Na⁺、Ca²⁺、Mg²⁺在 5 月急剧降低,之后 一直呈降低趋势,8月后浓度又逐渐上升,主要是受 到雨季降水稀释及至2010年4月30日起位于拉易 洞上游约200 m处的污水处理厂开始运营,集中处 理由城镇排放出的生活和生产污水,使地下河中各 离子浓度在2010年4月后开始出现显著降低;基 于同样的原因, HCO₃、Cl⁻、NO₃、SO₄²⁻ 也出现类 似的时间变化规律,但 NO; 、SO₄ 等阴离子相对于 其他离子波动较大. 从下游地下河出口小龙洞处 2010年7~12月的数据来看,除HCO; 离子外,各

里湖地下河部分测试数据1)

取样时间	采样点	电导/μS·cm ⁻¹	рН	水温/℃	溶解氧(DO)	Cl -/mg·L-1	SO ₄ -/mg·L -1	NO ₃ -/mg·L ⁻¹
	拉易洞	347	7. 73	20. 4	——————————————————————————————————————	8. 62	73. 84	4. 16
2010-05					_			
	凉风洞	392	7. 64	20. 1	_	9. 48	68. 49	1. 15
	拉易洞	419	7. 11	21	_	7. 31	61. 3	8. 37
	桥村	396	7. 19	19. 5	_	4. 47	42. 1	12. 36
2010-10	凉风洞	387	7. 54	20. 5	_	4.61	58. 9	18. 51
2010-10	红星河出口	350	7. 93	20. 1	_	2.79	56. 6	8. 68
	干田坝	380	8.08	20. 6	_	4. 23	54. 5	16. 96
	小龙洞	322	7. 48	20. 4	_	0.74	16. 2	5
	南丹河	365	7. 69	18. 7	7. 74	2. 56	45. 35	15. 34
	拉易河	367	7. 55	19. 5	7. 12	8.62	47. 11	14. 52
	桥村水源地	311	7. 24	18.8	6. 88	2.3	21. 59	7. 19
2014-05-16	凉风洞	349	7.63	20. 8	7. 9	3.66	45	12.04
	红星河出口	295	7.86	19. 5	8. 64	2. 27	38. 15	7. 19
	甘田坝	352	7. 99	20. 1	8. 76	3.49	44. 35	11.58
	小龙洞	364	7. 63	20. 9	8. 62	1.45	16. 58	3. 97
	南丹河	359	8. 87	24. 6	13. 55	5. 63	48. 1	4. 04
	拉易河	412	7.72	22. 4	9. 67	10.44	46. 5	9. 1
	桥村水源地	419	7. 1	19. 5	5. 16	4.51	40. 26	11.47
2014-10-18	凉风洞	373	7.51	20. 9	7.48	6.65	40. 38	14. 88
	红星河出口	368	7. 6	20. 1	5. 15	3.42	27. 92	8. 09
	甘田坝	373	7. 99	20. 5	8. 38	6. 11	40. 93	13. 98
	小龙洞	337	7.47	21.7	8.6	0.97	14 24	4 27

1)"一"表示数据缺省;部分数据引自文献[23]

离子浓度均较上游和中游的拉易洞和凉风洞低,且 基本保持在较低水平,随时间变化小.

3.3 里湖地下河硫同位素、氮氧同位素组成

测得里湖地下河 2014 年 5 月水体中 δ^{34} S-SO₄² 值在 - 4.12‰ ~ - 0.93‰ 之间波动, 平均为 -2.46‰(表2); δ¹5N-NO₃ 值在 4.79‰ ~ 10.22‰ 之间, 平均为 7.71‰, δ¹⁸O-NO₃ 值在 5.62‰ ~ 21.76‰之间,平均为12.53‰. 同年10月的硝酸盐 中的氮氧同位素组成与 5 月相比,δ¹⁵N-NO₃ 值变化 幅度更大, 在 0.26‰~11.58‰之间, 平均为 7.50‰, δ¹δO-NO; 值与 5 月相比普遍偏低, 在 -2.33‰~12.56‰之间,平均为6.22‰.

表 2 2014 年里湖地下河硫同位素、氮氧同位素特征

T 11 2	Values of δ^{34} S in sulfur	1.1 1 C 9 15 N T	1 2 180	T 1 T 1 1	1
rabie 2	values of o 5 in sumur	and the values of o in a	and o O in murate in	Linu Lake underground	i water in the year 2014

取样时间	样点名称	$\delta^{34} S - SO_4^{2-} / \% o$	$\delta^{15} \text{N-NO}_3^- / \% \sigma$	$\delta^{18} \text{O-NO}_3^- / \% \sigma$	$\delta^{15}\text{N-NO}_3^-/\delta^{18}\text{O-NO}_3^-$
	南丹河	-0.93	4. 79	9. 13	0. 52
	拉易河	-3.02	6. 92	6. 03	1. 15
	桥村水源地	-1.12	8. 98	21. 76	0.41
2014-05-16	凉风洞	-4.08	7. 17	5. 62	1. 28
	红星河出口	-4.12	7. 73	12. 97	0.60
	甘田坝	-1.37	8. 21	11. 22	0.73
	小龙洞	-2.61	10. 22	21	0.49
	南丹河	1)	7. 83	12. 56	0.62
	拉易河	_	0. 26	-2.33	-0.11
	桥村水源地	_	11.58	3.66	3. 16
2014-10-18	凉风洞	_	9. 76	4. 87	2. 00
	红星河出口	_	8. 7	6. 96	1. 25
	甘田坝	_	9. 26	5. 52	1. 68
	小龙洞	_	5. 1	12. 32	0.41

1)"一"表示数据缺省

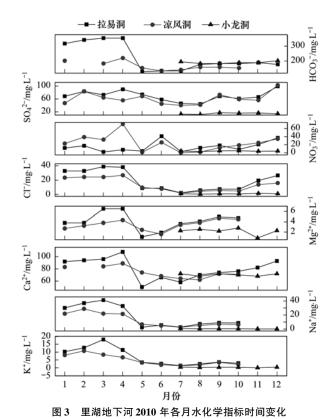


Fig. 3 Temporal variation of the hydrochemistry in Lihu Lake underground water in each month of the year 2010

4 里湖地下河硫酸盐、硝酸盐来源研究

4.1 里湖地下河硫酸盐和硝酸盐时空变化特征

里湖地下河中 SO_4^{2-} 较高,2010 年 5 月平均浓度为 71.17 $mg \cdot L^{-1}$,10 月平均浓度为 48.27 $mg \cdot L^{-1}$,2014年5月平均为 47.11 $mg \cdot L^{-1}$,10 月平均为 48.02 $mg \cdot L^{-1}$,远远高于世界河水 SO_4^{2-} 的平

均值 11.2 mg·L^{-1[26]}. 图 4 对比了里湖地下河 2010 年 5 月、10 月与 2014 年 5 月、10 月的 SO₄²⁻ 浓度时空变化,其中 2010 年大于 2014 年,表明从 2010 年污水处理厂开始运营后,虽其浓度仍偏高,但比 2010 年有所降低. NO₃⁻ 浓度在 1.15 ~ 71.21 mg·L⁻¹之间,其时间变化表现出不同于 SO₄²⁻ 的特征. 从图 4 可以看出 2014 年 5、10 月的 NO₃⁻ 浓度 都分别大于 2010 年 5、10 月,这说明硝酸盐的污染在加剧,指示了工、农业活动的加剧和城市污水的排放增加了地下河的氮负荷. 从空间分布上看,上游南丹河,拉易洞、中游凉风洞、甘田坝等硫酸盐、硝酸盐浓度较高,下游小龙洞处则较低. 结合实地考察,在城关镇小场采集的南丹河水样流经南丹县

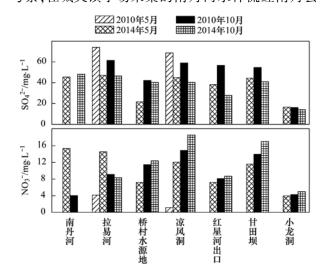


图 4 里湖地下河硫酸盐、硝酸盐时空分布变化

Fig. 4 Spatial and temporal variation of sulfur and nitrate in Lihu Lake underground water

城,途经居民区,菜市场,沿途生活、生产废水直接或间接地排放到南丹河,污染物质在向下运移的途中不断积累;拉易洞为地表河转为地下河的入口,汇集了来自上游的大量地表污水,加上周围分布有采石场、水泥厂、砖厂等,堆积在地表的岩石风化物和工业废水被输送到河水中;凉风洞周围正在进行旅游开发,地下河污染负荷随人类活动的加剧而增大;甘田坝周围为大片农田,农业活动也是地下水污染来源之一.据此粗略地认为地下水中硫酸盐、硝酸盐主要是来源于人类活动,但具体的污染来源需要结合硫同位素、氮氧同位素进行示踪.

4.2 δ³⁴S-SO₄²⁻ 特征及其 SO₄²⁻ 来源

地下水中硫酸盐主要有以下几种来源:大气降水、蒸发岩的溶解、硫化物的氧化及土壤带中有机硫的矿化,海岸地区还受到海水入侵的影响或传输的海盐通过降水补给而导致地下水中硫酸盐浓度增大^[27].此外,随着工农业的发展和城市化进程的加快,人类活动(如开采矿床、施用化肥等)的输入也成为硫酸盐的主要来源之一^[28~31].稳定同位素的组成(8³⁴S)代表各污染源的特征化学指纹,不同来源的8³⁴S存在较大差异,这使利用硫稳定同位素示踪污染来源成为可能^[32],而被广泛运用于自然生态系统中各种硫源的示踪和地球化学循环的研究^[33].

里湖地下河中δ³⁴S-SO₄² 值在 - 4.12‰ ~ -0.93‰之间,平均值为 -2.47‰(表 2),其相对较窄的分布范围反映了当地硫源较为单一.里湖地下河流经地区出露的二叠纪、三叠纪地层中偶夹薄层石膏,地下水在流经石膏层时,水岩作用的结果是水体会有较高的 SO₄² 浓度和高的δ³⁴S值.临近地区贵州的海相蒸发岩(石膏)的δ³⁴S值的变化区间为23.7‰ ~ 29.56‰ [³⁴],四川盆地三叠系硬石膏δ³⁴S值为14.7‰ ~ 35.4‰ [³⁵]. Querol等 [³⁶]研究西班牙地区地下水、地表水及矿物中 SO₄² 的硫同位素组成时,发现三叠纪和中新世石膏的δ³⁴S值多数分布在12‰ ~ 14‰之间.通常地层中石膏的δ³⁴S值均偏正,与研究区地下水中δ³⁴S值 - 4.12‰ ~ -0.93‰之间差异较大,可以得出里湖地下河中没有或较少存在石膏溶解的情况.

据 Moncaster 等^[37] 报道, 英国化肥的 δ^{34} S 值在 5.4% ϵ^{38} 8.5%之间; Mizota 等^[38] 报道了日本的化肥 硫铵 (俗称肥田粉) 和普通过磷酸钙 δ^{34} S 值为 10.2%, 在澳大利亚和新西兰的更高, 在 15.7% ϵ^{38} 20.7% 之间; 在四川盆地西南部的峨眉山, 人造化肥为 7.1% ϵ^{38} 8.9% ϵ^{38} 1.20 由凉风洞以下的里湖中下

游地区虽分布有大片农田,但农业化肥的8³⁴S值皆分布在正值范围内,与里湖地下水的8³⁴S-SO₄² 值主要是在负值范围不一致,由此认为农业活动中施用的化肥不是里湖地下河 SO₄² 的主要来源.

里湖地下河的补给来源主要是大气降水,所 属的河池市是广西地区酸雨较严重的城市之一, 约82%的大气降水的 pH 值 < 5.6^[39],酸雨可向地 下河中输入大量 SO₄². 据广西地区的柳州气象局 地面站、桂林市环境保护监测站、合山电厂化学 分场等的监测数据,湘桂走廊地区大气降水的 δ^{34} S- SO_4^{2-} 值的变化范围为 - 4.8‰ ~ - 0.1‰,集中分 布在负值范围内[40],涵盖了里湖地下水的硫同位 素组成范围(-4.12%~~-0.93%),据此推断地 下水中 SO₄²⁻ 大部分来自于大气降水. 又将里湖地 下河的 δ^{34} S-SO₄² 值与 1/SO₄² 做图(图 5), 水体 $δ^{34}S-SO_4^{2-}$ 值主要落在大气降水的 $δ^{34}S$ 值范围内,部 分落在大气降水和煤重合的值域内,但里湖流经 地层主要为石炭系下统岩关阶、石炭系上统马平 群、二叠系下统栖霞阶、二叠系上统、三叠系下 统,三叠系中统百逢组,并没有煤系地层的存在, 出现这种情况可能是由于煤炭是里湖地区工业生 产和居民生活的主要能源,洪业汤等[41]研究中国 大气降水的南北分异时发现,中国在长江以南地 区,由拉萨,经贵阳往深圳地区,大气降水的δ³⁴S值 均为负值,这与中国煤炭δ³⁴S值的南北分异相似, 认为煤炭燃烧过程中的同位素分馏效应造成了这 种分异.

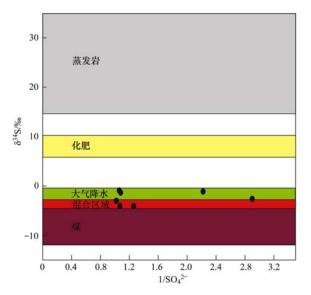


图 5 里湖地下水 δ^{34} S与 $1/SO_4^{2-}$ 的变化关系

Fig. 5 Variation relationship of $\delta^{34}\! S$ and $1/SO_4^2$ - in the Lihu Lake underground water

4.3 δ¹⁸O-NO₃ 和δ¹⁵N-NO₃ 特征及其 NO₃ 来源

4.3.1 反硝化作用的识别

地下水中的硝酸盐来源很多,而且一经污染则 难以治理,因此查明硝酸盐来源,对从根源上减少硝 酸盐的输入具有重要意义[42]. 岩溶地区的土壤、岩 石和包气带的渗透性较好,由源进入地下水过程中 氮同位素分馏作用通常较小^[43~45],应用δ¹⁵N-NO₃, 并结合δ¹⁸O-NO; 能够有效地进行地下水污染溯源. 但发生反硝化作用导致氮同位素分馏,致使不同氮 源的硝酸盐同位素组成值域发生重合的可能仍然存 在,因此,有必要辨识反硝化过程,排除同位素分馏 导致的氮源识别干扰[46]. 在识别反硝化作用之前 还应先判定该区的地下水环境是否适宜反硝化作用 的发生. 由于反硝化作用发生在厌氧环境下,因此 水中溶解氧的多少可以作为判别指标之一,研究发 现在 DO 浓度 < 0.2 mg·L⁻¹条件下,反硝化速率最 理想[47]. 而 Gillham 等[48] 经野外调查研究认为,地 下水环境中反硝化作用的 DO 上限为 2.0 mg·L⁻¹; Desimone 等[47]对地下水NO, -N污染晕监测分析时 发现,在 DO 浓度为 2~6 mg·L⁻¹条件下仍有反硝化 作用的存在,但速率很小[49]. 反硝化作用是受 NO3 污染地下水的一种重要的自净过程,可以降低水中 NO、的污染水平,一般反硝化过程中,NO、浓度降 低伴随着δ¹⁵N/δ¹⁸O成比例增加,增加比率接近2: 1^[50],应用这种线性关系,可以识别地下水是否发生 反硝化作用.

据 2014 年 5 月和 10 月现场测定的 DO 数据 (表1),仅10月在桥村水源地测得的DO值为5.16 mg·L⁻¹,小于 6 mg·L⁻¹,其余样点的 DO 值在 6.88 ~13.55 mg·L⁻¹之间,平均为7.92 mg·L⁻¹,指示当 地地下水环境并不适宜反硝化作用的进行. 从研究 区的水文地质条件来看,南丹河到甘田坝的里湖中 上游段,地下水埋藏较浅,小于30 m,而且地下河管 道为很大的洞穴,明暗流交替,空气充足,属于好氧 环境,硝化作用占主导地位,再次印证了以上推断. 地下河大多数样点δ¹⁵N在 4.79‰~11.58‰这一较 小的变化范围,相反,δ¹⁸O却有较广的分布范围,在 -2.33‰ ~ 21.76‰间. 通过计算δ¹5N/δ¹8O得到的 比值多数在 -0.11~1.68 之间,比值小于2,仅采于 2014年10月的桥村水源地与凉风洞比值较大,分 别为 3. 16 和 2. 00(表 2). 从图 6 也可以看出,δ¹⁵N 与 NO、的浓度间并不存在显著的相关关系,并未出 现 NO、浓度降低伴随着δ¹⁵N增加这一反硝化作用 的线性关系. 综上分析,判断反硝化作用并不是影

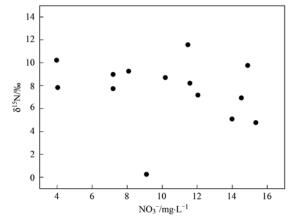


图 6 里湖地下水 NO_3^- 浓度与 $\delta^{15}N$ 的关系

Fig. 6 Relationship between the NO_3^- concentration and the $\delta^{15}N$ values

响同位素变化的因素.

4.3.2 地下水硝酸盐来源

氯在自然界中是相对稳定的元素,其可能来源包括农用钾肥,动物粪便,生活污水等,因此氯可以作为指示污染源的元素^[51]. NO_3^-/Cl^- 可以用来识别 NO_3^- 浓度变化过程中的稀释效应和生物作用^[52,53]. 2014年5月,里湖地下河中 Cl^- 的平均浓度为 2. 97 $mg\cdot L^{-1}$,10月平均为 5. 39 $mg\cdot L^{-1}$,受雨水稀释作用明显,从图 7可以看出,大多数采样点的 NO_3^- 与 Cl^- 呈一定的正相关趋势,指示它们具有相似的来源,5月的样品中 NO_3^- 与 Cl^- 之间呈正相关($R^2=0.4610$),这揭示样品 NO_3^- 受到大气降水和其他来源的混合作用.

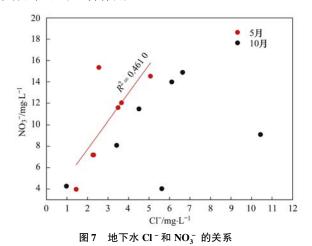


Fig. 7 Relationship between Cl $^-$ and NO $_3^-$ concentrations in Lihu Lake underground water

地下水 δ^{15} N-NO₃⁻ 值在 0. 26‰ ~ 11. 58‰之间, 平均为 7. 61‰, δ^{18} O-NO₃⁻ 值在 - 2. 33‰ ~ 21. 76‰ 之间,平均为 9. 38‰. 图 8 为较常用的不同来源的

硝酸盐的 N、O 同位素的典型特征值,将里湖各采 样点的 N、O 同位素在图 8 中表示,主要是落在土 壤氮、人畜粪便和污水的交叉区间,说明人类生活 排放的污水、饲养家畜排出的粪便以及有机氮浓度 较高的土壤是当地硝酸盐主要来源. 一般来说,土 壤 N 的浓度较为稳定,人畜粪便和生产生活污水排 放是引起里湖流域硝酸盐浓度在空间上波动变化的 主要原因. 需要特别注意的是,在2014年5月采集 的桥村水源地和小龙洞这两个点的 N、O 同位素主 要落在大气沉降 NO; 这一范围内,桥村水源地为当 地的备用水源,居民注重对饮水水质的保护,小龙洞 周围为大片的未开垦的山地,植被覆盖良好,远离人 类活动,且这两个采样点的 NO; 浓度显著低于其他 采样点(图4),这都说明在化肥,人畜粪便和污水等 输入较少,森林覆盖条件较好的地区,雨季大气降水 的混入也是当地地下水重要的氮源[54~56],且对样品 N 同位素影响较大; 在旱季的 10 月份, 大气降水较 少,土壤氮、人畜粪便和污水便成为了主要的污染 来源.

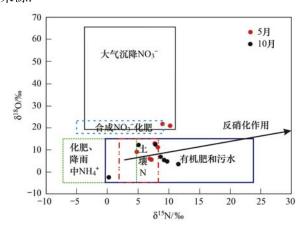


图 8 不同来源硝酸盐的δ¹⁵N和δ¹⁸O典型值^[57]

Fig. 8 Typical ranges of $\delta^{15}N$ and $\delta^{18}O$ values of nitrate of different sources

5 结论

- (1)里湖地下河水中主要阴离子为 HCO₃⁻,主要阳离子为 Ca²⁺、Mg²⁺,基本水化学类型为 HCO₃-Ca型. 水体中离子的主要来源为碳酸盐岩的风化,其次各离子变化还受到人类活动的影响,除 HCO₃⁻外,其他离子浓度在里湖地下河上游、中游偏高,下游偏低. 受中亚热带季风气候的影响,地下水中各离子浓度呈现出雨季降低,旱季增大的特点.
- (2)自2010年污水处理厂开始运营后,地下河中硫酸盐浓度有所降低,但仍然偏高,硝酸盐浓度略有增加,指示当地硝酸盐污染在加剧.从空间变化

- 上看,上游南丹河,拉易洞、中游凉风洞、甘田坝等 人类活动强度较大,地下河硫酸盐、硝酸盐浓度较 高,下游小龙洞处植被良好且远离人类活动干扰,其 硫酸盐、硝酸盐浓度较低.
- (3)里湖地下河 δ^{34} S-SO₄² 值分布在 -4.12‰ ~ -0.93‰这一相对较窄的范围,反映了当地硫源较为单一,并且该值域与大气降水的 δ^{34} S值一致,因此认为大气降水输入是当地 SO₄² 的主要来源.
- (4)将2014年与2010年相对比,里湖地下河中硝酸盐浓度呈增加趋势,表明硝酸盐污染在加剧.根据地下水 DO 值,并结合 NO_3^- 与 $\delta^{15}N$ 、 NO_3^- 与 Cl^- 、 $\delta^{15}N-NO_3^-$ 与 $\delta^{18}O-NO_3^-$ 的关系排除了反硝化作用对氮氧同位素的影响.地下水硝酸盐的 $\delta^{15}N$ 值在 0.26%。~11.58%。之间, δ^{18} O值在 ~2.33%。~21.76%。之间,指示土壤氮、人畜粪便和污水是研究区硝酸盐的主要贡献者,为有针对地开展硝酸盐污染防治提供了依据.

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