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柳江盆地浅层地下水硝酸型水特征和成因分析

徐进1,何江涛1*,彭聪2,曾颖3

(1. 中国地质大学(北京)水资源与环境学院, 北京 100083; 2. 中国地质科学院岩溶地质研究所, 桂林 541004; 3. 江西省建筑设计研究总院, 南昌 330000)

摘要:近年来地下水硝酸盐污染状况越来越严重,人类活动对地下水的影响加剧,不仅造成污染,甚至改变了地下水的水化学类型,出现了硝酸型地下水.本研究以柳江盆地为例,分析了该地区浅层地下水中硝酸型水的特征和影响控制因素,探讨了硝酸型水的研究意义.结果表明,柳江盆地浅层地下水中硝酸盐污染严重;在全区211组有效数据中,硝酸型水占比20.9%,主要分布在研究区东南部,北部山区分布较少且零散;根据硝酸型水的特征可将其划分为高TDS硝酸型水和低TDS硝酸型水,高TDS硝酸型水的TDS和总硬度浓度均较大,且浓度范围较广,低TDS硝酸型水的TDS和总硬度浓度值和浓度范围均较小;硝酸型水的形成主要受人类生活污水、农业施肥、化粪池和垃圾渗滤液下渗等影响,其中,高TDS硝酸型水的污染负荷通常高于低TDS硝酸型水;通过对硝酸型水的分析研究,能够尽早发现污染并不严重但已有污染趋势的区域,以便及时进行地下水的污染防治,避免水质进一步恶化.

关键词:柳江盆地; 地下水; 硝酸型水; 因子分析; 成因分析; 总溶解性固体(TDS)

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Characteristics and Genesis of NO₃ Type Water in Shallow Groundwater in Liujiang Basin

XU Jin¹, HE Jiang-tao^{1*}, PENG Cong², ZENG Ying³

(1. School of Water Resources and Environment, China University of Geosciences (Beijing), Beijing 100083, China; 2. Institute of Karst Geology, China Academy of Geological Science, Guilin 541004, China; 3. Jiangxi Province Architectural Design & Research Institute, Nanchang 330000, China)

Abstract: In recent years, groundwater nitrate pollution has become more and more severe. Anthropogenic activities have become more significant, not only causing pollution, but also changing the hydrochemical type of groundwater and leading to the emergence of NO₃ type groundwater. This study focuses on the Liujiang Basin. The characteristics and influencing factors of NO₃ type water in shallow groundwater are analyzed, and the significance of NO₃ type water is discussed. Results show that shallow groundwater in the Liujiang Basin is of poor quality and nitrate pollution is severe. NO₃ type water comprises 20.9% of all effective data for the area and is mainly distributed in the southeast and a few parts of the northern mountainous area. Based on the characteristics of NO₃ type water, this can be classified as high TDS NO₃ type water or low TDS NO₃ type water. TDS and total hardness concentration values of high TDS nitrate type water are high and the concentration range is wide. Conversely, TDS and total hardness concentration and the concentration range of low TDS NO₃ type water are both low. The formation of NO₃ type water is mainly driven by human domestic sewage, agricultural fertilization, and septic tank and landfill leachate infiltration, among others. The pollution load of high TDS NO₃ type water is generally higher than that of low TDS NO₃ type water. Analysis of NO₃ type water makes it possible to prevent and control groundwater pollution when there is an increasing pollution trend but pollution is not yet severe.

Key words: Liujiang Basin; groundwater; the NO₃ type water; factor analysis; cause analysis; total dissolved solid (TDS)

近年来,国内外地下水污染状况日趋严重,尤其是地下水硝酸盐污染. 国外关于硝酸盐超标的报道已较多^[1~4],主要的输入源是化粪池以及生活污水渗漏^[5~8];国内硝酸盐污染以北方为主^[9~11],这主要是养殖、施肥、居民生活污水以及垃圾粪便下渗等人类活动使得大量的氮输入到地下水中造成的^[12~18]. 人类活动在造成地下水污染的同时,也影响到了地下水的水化学类型^[19],国内已有陆续报道,在一些地区地下水中出现了一种新的水化学类型,即硝酸型水.

所谓硝酸型水,一般指硝酸根在阴离子中的毫克当量百分数大于25%,成为影响到水化学命名的主要阴离子组分的地下水.2006年,苑丽华^[20]发现松嫩平原长期以来的农业生产使用化肥造成地下水中"三氮"含量逐年升高,水化学类型中出现了硝

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作者简介: 徐进(1995~), 男, 硕士研究生, 主要研究方向为地下水环境及水文地质, E-mail:1589534612@ qq. com

^{*} 通信作者,E-mail:jthe@cugb.edu.cn

酸型水. 2014 年周迅等^[21]在对福建省晋江市的研究中也发现了硝酸型水,且在晋江地区硝酸型地下水均有偏酸性、低矿化度等特征. 2016 年,朱亮等^[22]在研究东胜城区及周边地区浅层地下水硝酸盐分布及影响因素的过程中发现,研究区地下水硝酸盐超标率较高达21%,且超标点主要分布在人类活动强度较大、水位埋深较浅的地区,局部地区出现硝酸型地下水.

尽管在以往的调查评价中,已经频频出现硝酸型水类型,但是对于硝酸型水的特征、成因及其研究意义等方面还不够深入.柳江盆地浅层地下水是当地居民的主要饮用水源,地下水的污染将直接危害到居民的身体健康^[23].因此,本文以柳江盆地为例,进一步探讨硝酸型地下水的分布特征、主要影响因素和其研究意义,以期为硝酸型地下水的深入认识提供一定的研究基础.

1 研究区概况

1.1 自然地理及水文地质概况

柳江盆地位于河北省秦皇岛市抚宁县境内,以石门寨为中心,位于秦皇岛北28 km 处.研究区主要人口聚居区为石门寨镇、驻操营镇,其余地区多为村落.柳江盆地多年平均降水量为698.5 mm,多年平均蒸发量为1600 mm.降雨多集中在6~9月,多暴雨,雨量年际变化及季节变化较大.

柳江盆地地处燕山山脉东段,濒临渤海湾、华北平原,长约 15 km,宽约 12 km,总面积约 180 km².柳江向斜是燕山运动期,由于华北板块从北西方向向太平洋板块仰冲挤压形成,导致向斜西翼较陡,东翼较缓,中间低缓,大石河从中流过.地势北高南低,西高东低.北部和西部是平均海拔在200~300 m之间的低山区,东部为海拔在160~300 m之间的丘陵区,盆地中部主要为低缓丘陵,分布有大石河冲积平原.柳江盆地地表水系较为发育,主要以大石河流域为主,自北向南流经本区并最终注入渤海,区内地下水分水岭与地表水一致.

柳江盆地是一个不对称的向斜盆地,低山丘陵将盆地分割成近南北向的山间河谷.区内主要有两大含水层.松散岩类孔隙水主要分布在大石河主、支流的河谷中,石门寨、驻操营等地主要分布大面积的第四系潜水含水层.碳酸盐岩类裂隙岩溶水在大石河主、干及主要支流的河谷分布,河谷两侧丘陵区域少量分布.柳江盆地第四系孔隙潜水和碳酸盐岩裂隙岩溶水之间水力联系密切.盆地四周及基

底为早远古绥中花岗岩、燕山期花岗岩、斑状花岗岩等,构成弱透水层.由于花岗岩构成了盆地四周的相对隔水边界,使柳江盆地构成了一个完整封闭的储水构造(图1).

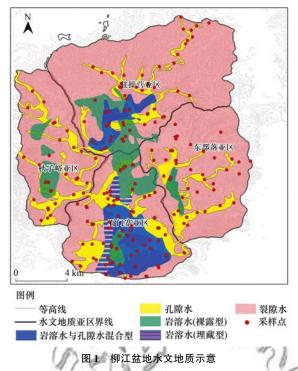


Fig. 1 Hydrogeology map of the Liujiang Basin

柳江盆地地下水的主要补给来源有:降雨渗入补给、河流的渗入补给、灌溉回归水的渗入补给等.研究区内基岩裸露,植被稀疏,河谷地带砂卵石层松散,具有良好的入渗条件,使得地表水、地下水水力联系密切.由于柳江盆地内三面地势较高,所以地表以及地下水主要的运移方向是由四周向盆地中间运动汇聚,然后在盆地东南出口得到排泄.

根据大石河的汇水条件以及研究区的地质构造、岩石类型、地下水分水岭可将研究区分为四个水文地质单元,大石河中游石门寨汇水区、东宫河东部落汇水亚区、大石河上游驻操营汇水亚区和大石河上游秋子峪汇水亚区.

1.2 浅层地下水水质特征

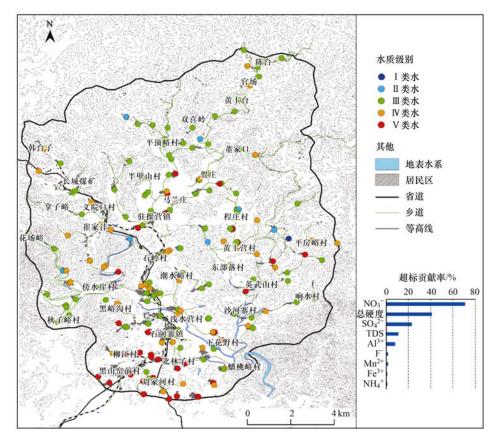
本文中的地下水分析数据来自于文献[24],分别在2013和2014年完成了137组简分析和149组全分析的水样,其中包含有21组地表水水样,汇水区外30组水样,区内地下水检测235组水样.简分析主要测定了钾、钠、钙、镁、氨、重碳酸根、碳酸根、氯离子、硫酸根、氟、硝酸根、亚硝酸根、溶解性总固体、矿化度、游离二氧化碳、总硬度、永久

硬度、暂时硬度、负硬度、总碱度、总酸度、pH值,全分析在简分析的基础上增加了铝、铁、偏硅酸、锰、铜、锌、汞、铅、镉、铬、砷、溴化物、碘、磷酸盐、耗氧量.样品采集后立即用封口胶密闭封存,并储存于低温保温箱中,均送至中化地质矿山总局河北地质勘查院,按照国家相关标准进行测试.采样点布置与采集遵循文献[25]相关要求.

为了保证研究数据的代表性和可靠性,在保证 采样与分析测试的标准规范基础上,对已获取的地 下水数据的区域代表性和可靠性进行筛选和检验. 在已有数据的基础上,剔除区外、地表和深井水 样,以及平行样和电荷不平衡、碳酸不平衡等不可 用水样,最后得到柳江盆地浅层地下水有效分析数 据为211组,采样点位置见图1. 研究区最主要的水化学类型为 $HCO_3 \cdot SO_4 - Ca \times SO_4 \cdot HCO_3 - Ca \times HCO_3 - Ca \times SO_4 - Ca \times SO_4 \cdot Cl - Ca$. $HCO_3 \cdot SO_4 - Ca$ 型水分布最广,主要分布在研究区四周山区, $HCO_3 - Ca$ 型主要分布在盆地中东部分, $SO_4 \cdot HCO_3 - Ca$ 型分布较为分散, $SO_4 \cdot Cl - Ca$ 型水主要分布在盆地南部,分布面积相对较小.

研究区主要以Ⅲ类水为主,约占48%. 其次是Ⅳ类和Ⅴ类水,分别占25%和21%. Ⅰ类水和Ⅱ类水均较少,分别占1%和5%,见图2.

从图 2 中可以看出 NO₃ 的贡献率远大于其他指标,说明柳江盆地地区硝酸盐超标严重. 另外也可以看出,水质较差的Ⅳ类和Ⅴ类水主要分布在石门寨南部,在秋子峪亚区东南部、东部落亚区中东部、驻操营南部亚区等区域少量分布.



参评指标: $Na^+ \ SO_4^{2-} \ Cl^- \ NO_3^- \ NO_2^- \ NH_4^+ \ Pb^{2+} \ Zn^{2+} \ As^{3+} \ Hg^+ \ 总硬度 \ TDS \ pH \ F^- \ COD \ Fe^{3+} \ Mn^{2+} \ Al^{3+} \ Cd^{3+} \ 91 \ 项; 评价方法: 文献[26] 中指标的限值及推荐的单指标最大类别方法$

图 2 研究区地下水质量评价分级[24]

Fig. 2 Groundwater quality classification map of the study area

2 硝酸型水特征及成因分析

2.1 硝酸型水的特征

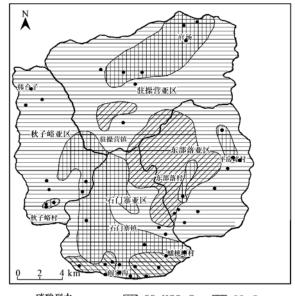
根据硝酸型水的定义重新计算研究区浅层地下水的水化学类型,可以得出研究区硝酸型水的类型

及数量. 计算统计结果显示, 研究区 211 组浅层有效地下水样品中, 共有 44 组水化学类型为硝酸型, 占 20.9%(图 3), 主要分布在盆地东南部, 东部落亚区东部和秋子峪亚区东部少部分分布, 其他区域分布较为分散, 整体上分布在研究区四周山区. 硝

酸型水的类型包括 7 类, 分别为: HCO₃·NO₃-Ca 型水 9 组, NO₃-Ca 型水 5 组, NO₃·HCO₃-Ca 型水 5 组, NO₃·SO₄-Ca 型水 13 组, SO₄·NO₃-Ca 型水 8 组 SO₄·HCO₃·HNO₃-Ca 型 3 组, HCO₃·SO₄·HNO₃-Ca 型 1 组.

为进一步分析硝酸型水的特征,分析硝酸盐含量和硝酸型水的关系,做出硝酸盐的绝对含量和相对含量的关系(见图 4). 其中,相对含量以 25%进行分界,绝对含量以 88.0 mg·L⁻¹进行分界,将所有数据点分为四类,对应图 4 中的 4 个区.进行分区之后,可以看出,①区的水样点是超Ⅲ类水,但不是硝酸型水,②区的水样点既是超Ⅲ类水也是硝酸型水,③区的水样点既不是超Ⅲ类水也不是硝酸型水,④区的水样点是硝酸型水但不是超Ⅲ类水.

从整个研究区的分布情况来看(图 5), ③区的水样点最多, 分布最广, 其次是②区的水样点, 最后是①区和④区的水样点. ①区的水样点主要分布



SO₄-Ca
HCO₃-SO₄-Ca

图 3 硝酸型水分布示意

Fig. 3 Distribution of NO₃ type water

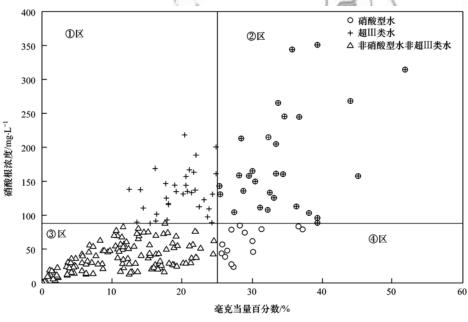


图 4 硝酸根浓度与毫克当量百分数关系

Fig. 4 Nitrate concentration and mEq percentage relationship diagram

在驻操营亚区南部,以及整个研究区南部区域,另在东部落亚区小面积分布,除研究区南部分布的点之外,其他地区的点都处于地势较平的丘陵区;②区主要的分布区域是研究区最南端,东部落东部一带小区域分布,其他区域零星分布,整体上②区的水样点分布在四周山区,盆地中心平原区几乎无②区的水样点;③区的水样点数量较多,分布较广;④区的水样点较少,且分布较为零散,但都分布在研究区边界山区中,TDS含量普遍较低.

分区之后,对各个分区内主要指标进行统计对比,见图 6. 从中可以出:①区,TDS 和总硬度是 4个分区中最高的,硝酸盐浓度超标;②区 TDS 和总硬度很高,硝酸盐浓度是 4个分区最高的,浓度范围也最大;③区 TDS 和总硬度较低,硝酸盐浓度是 4个分区最低;④区 TDS 和总硬度最低,硝酸盐浓度较高.对比①区和②区各指标数据,②区的 TDS 较①区低,但硝酸盐浓度②区比①区高出很多,③区和④区也有类似的特征,也就是说硝酸型水具

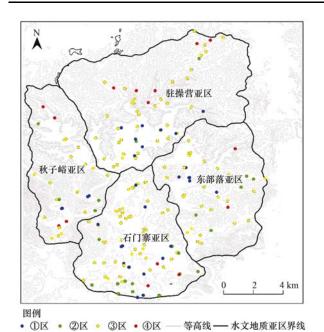


图 5 分区后各区水点的分布情况 Fig. 5 Distribution of each subarea after partition

有 TDS 和总硬度偏低等特征.

学

为了更好地总结出硝酸型水的特征, 做出硝酸 型水点的硝酸根浓度和 TDS 浓度关系, 并区分出② 区和④区的水点(图7). 从图7中看出,硝酸型水 的硝酸根浓度和 TDS 有良好的线性关系, ②区为高 TDS的硝酸型水, ④区为低 TDS的硝酸型水. 高 TDS 硝酸型水的硝酸根浓度值大致在 90~350 mg·L-1的范围内,浓度值和浓度范围均较大,低 TDS 硝酸型水的硝酸根浓度大致在 50~90 mg·L-1 之间. 这一现象说明, 高 TDS 的地下水形成硝酸型 水往往需要较高的硝酸盐污染负荷, 而低 TDS 的地 下水形成硝酸型水则不需要太高的硝酸盐污染负 荷. 对于一般地下水而言, 硝酸盐的含量通常较 低,在天然状态下难以形成硝酸型水.但是在人类 活动影响下, 硝酸盐污染加剧即会出现硝酸型水. 需要特别引起关注的是本研究中所出现的低 TDS 硝酸型水,虽然硝酸盐含量未超标,但是已形成了

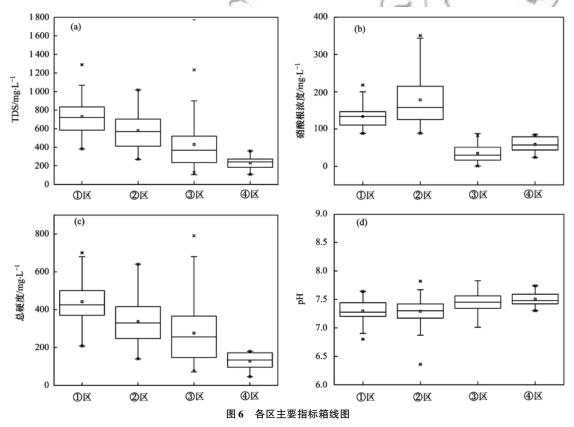


Fig. 6 Box plots showing major parameters

硝酸型水,说明这些地区地下水水质已在人类活动影响下开始恶化,所以硝酸型水的引入能够突出硝酸盐浓度未超标,但是已受人类活动影响、有水质恶化趋势的区域,这样能够尽早发现污染区域和趋势,及时做出防范措施,避免水质进一步恶化.

2.2 硝酸型水的成因分析

2.2.1 因子分析

为探讨研究硝酸型水形成原因,本研究使用因子分析法分析柳江盆地浅层地下水的影响和控制因素,选取主要离子 K^+ 、 Na^+ 、 Ca^{2+} 、 Mg^{2+} 、 HCO_3^-

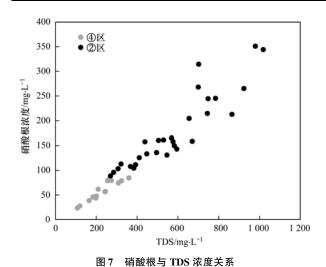


Fig. 7 Nitrate and TDS concentration diagram

(含 CO_3^{2-})、 SO_4^{2-} 、 CI^- 、 NO_3^- 、TDS、 NO_2^- 、氨和总硬度等指标. 利用因子分析法时取特征值大于 1, 旋转迭代的最大方差法.

因子分析法总共提取出3个主要因子(分别记为F1、F2、F3),解释的方差累积贡献率是78.78%(表1),代表了211组数据的绝大多数信息.F1的主要正得分变量是钠、钙、镁、重碳酸根、硫酸根、溶解性总固体、总硬度等变量,无得分为负的变量,表明F1代表的是含水层的溶解盐类水平,主要受天然水岩作用影响;F2主要的正得分变量是钾、氯离子、硝酸根,说明硝酸根离子和氯离子与钾离子有相似的来源途径,而氯离子主要来源于人类活动生产和生活污水排放,钾离子也可来源于人类农业施肥等活动,所以F2代表的是人类活动的影响;F3主要的正得分变量是亚硝酸根和氨,反映

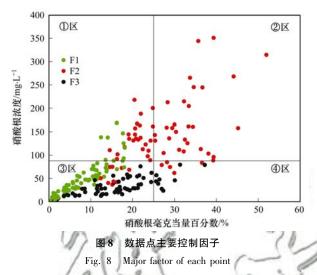
表1 旋转成份矩阵

Table	1 Rotate com	Rotate components matrix	
指标	F1	F2	F3
钾	0. 136	0. 543	-0.005
钠	0. 786	0. 239	0.010
钙	0.854	0.324	-0.060
镁	0.840	0. 203	-0.028
重碳酸根	0. 787	0.024	-0.007
氯离子	0. 293	0.897	0. 014
硫酸根	0. 920	-0.086	- 0. 044
TDS	0. 907	0. 392	-0.026
硝酸根	0. 096	0.938	-0.070
总硬度	0. 920	0.310	-0.054
亚硝酸根	-0.058	0.002	0. 919
氨	-0.025	-0.052	0. 920
特征值	6.076	1.712	1. 666
方差解释率/%	50. 630	14. 269	13. 884
累积方差解释率/%	50. 630	64. 899	78. 783

1) 黑体字表示各个指标在各控制因子中得分较高的因子

的是还原环境,主要是农村地区化粪池等渗漏污染 所影响.

在因子分析过程中,可得到每个数据点的因子得分情况,确定并统计每个数据点的主要控制因子,并在图 3 中区分 3 个主要控制因子(图 8).从图 8 中可以看出硝酸型水主要受 F2 影响,部分受F3 影响,其中高 TDS 硝酸型水均受 F2 影响,低TDS 硝酸型水中 6 个水点受 F2 影响,9 个水点受F3 影响.说明硝酸型水的形成主要是受人类活动污染输入控制.



2.2.2 相关性分析

为了进一步确定硝酸水的成因,进行硝酸根与 氯离子和钾离子的浓度相关性分析.

(1) 硝酸根与氯离子浓度相关性分析

做出硝酸根与氯离子浓度的关系图[图 9 (a)],两者相关性较好,对硝酸根与氯离子浓度进行拟合,拟合结果为 y = 2.41x - 1.717, R² 为 0.73,拟合效果较好,说明绝大多数点的硝酸根与氯离子相关性较好.硝酸型水水样点的硝酸根浓度和氯离子浓度线性关系明显[图 9(b)],相关性更好.无论是全区还是硝酸型水点,硝酸根浓度和氯离子浓度相关性都较好,说明在全区,硝酸根与氯离子有相同或相似的来源途径,且具有普遍性.而氯离子主要来源于生活污水垃圾渗滤液下渗等,与因子分析结果一致.进一步说明硝酸型水主要受控于生活污水或垃圾渗滤液下渗等.

(2) 硝酸根与钾离子相关性分析

首先做出硝酸根由于钾离子浓度关系图[图 10 (a)],发现数据点出现了两支的情况,一支硝酸根浓度均在钾离子浓度很低的区域,另一支,硝酸根浓度与钾离子相关性一般.硝酸型水水样点也有分

支的情况[图 10(b)]. 尽管硝酸根和钾离子之间没有很好的相关性, 但是从全区硝酸根浓度与钾离子

浓度的关系说明,硝酸型水还是受到了钾离子的影响,也就是说,农业施肥对硝酸型水有影响.

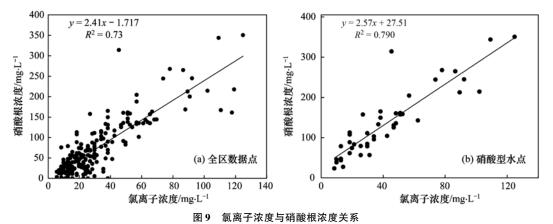


Fig. 9 Relationship between chloride and nitrate concentrations

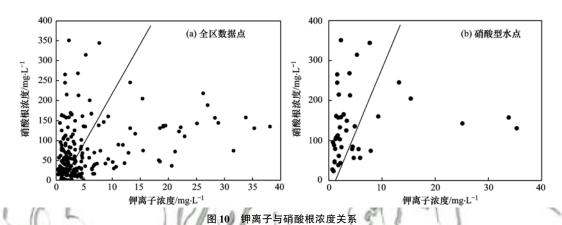


Fig. 10 Relationship between potassium and nitrate concentrations

总体而言,研究区硝酸型水主要受人类活动输入控制,受生活污水、农业施肥、化粪池和垃圾渗滤液下渗等因素影响,其中,高 TDS 硝酸型水的污染负荷通常高于低 TDS 硝酸型水.

3 结论

- (1)柳江盆地硝酸型水占比 20.9%,主要分布在盆地东南部,东部落亚区东部和秋子峪亚区东部少部分分布,其他区域分布较为分散,整体上分布在研究区四周山区;根据硝酸型水的特征可将其划分为高 TDS 硝酸型水和低 TDS 硝酸型水两类,高TDS 硝酸型水 TDS 浓度和总硬度浓度均较大,浓度范围较广,低 TDS 硝酸型水的 TDS 和总硬度浓度值和浓度范围均较小.
- (2)柳江盆地浅层地下水中硝酸型水的形成主要受人类生活污水、农业施肥、垃圾渗滤液下渗等影响,其中,高 TDS 硝酸型水的污染负荷通常高于低 TDS 硝酸型水.

(3)硝酸型水的提出,可以发现那些未污染但已有污染趋势的区域,尽早进行地下水的污染预防,以避免水质恶化,影响其使用.

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