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崇左响水地区地下水水质分析及健康风险评价

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摘要:以崇左响水地区为研究区域,对区域内 60 个地下水样品中的常规指标和金属元素进行测定和分析,运用内梅罗综合指数法分析了不同类型地下水环境质量,应用健康风险评价模型评价了不同类型地下水的健康风险.结果表明,井水、泉水和地下河水的常规指标和金属元素均出现不同程度的超标现象,水质级别均为较差级别,地下河水的综合评价分值(F = 4.26)最低,井水和泉水的综合评价分值(F = 7.10)相同. 高硬度和高矿化度利于 Cr 的富集,还原环境利于 As 的富集,Zn、Pb、Cd 和 Cu 经过的环境地球化学作用相似,Fe、Al 和 Mn 来源相似. 健康风险评价表明,3 种类型地下水的健康总风险偏高,大小顺序为井水 > 地下河水 > 泉水,主要来源于致癌性金属元素 Cr. 致癌总风险比非致癌总风险高 4~6 个数量级,致癌总风险均高于最大可接受风险水平(5.0×10^{-5} a $^{-1}$),非致癌总风险均小于可接受的健康风险水平(10^{-6} a $^{-1}$). 儿童健康总风险大于成人,经饮水途径引起的健康风险比皮肤接触途径高 2~3 个数量级. 从饮水安全考虑,在饮用前需对井水、地下河水和泉水进行适当处理并实施对 Cr 污染物的控制.

关键词:地下水;水质分析;内梅罗综合指数;健康风险评价;崇左响水地区

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Water Quality Analysis and Health Risk Assessment for Groundwater at Xiangshui, Chongzuo

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Abstract: To investigate the environmental quality and human health risks of different types of groundwater at Xiangshui, Chongzuo, several regular water quality indexes and concentrations of metals in 60 groundwater samples were measured and analyzed. The environmental quality of groundwater was analyzed by means of the Nemerow index. The health risks were assessed by using a human health risk assessment model. The regular water quality indexes and concentrations of metals of the well water, spring water, and underground river water exceeded the standards to different degrees. The environmental quality of groundwater was at a poor grade. The comprehensive evaluation score of underground river water (F = 4.26) was the lowest. The well water had the same score as spring water (F = 7.10). The high hardness and salinity were conducive to enrichment of Cr, and the reducing environment was of great advantage for the enrichment of As. The environmental geochemistry of Zn, Pb, Cd, and Cu was similar. The sources of Fe, Al, and Mn were similar. The results of the health risk assessment indicated that the health risks of well water, spring water, and underground river water were relatively high. The health risks decreased in the order of well water > underground river water > spring water. The health risks mainly came from the carcinogenic metallic element Cr. Carcinogenic risks were 4-6 orders of magnitude higher than noncarcinogenic risks. Carcinogenic risks were higher than the maximum allowance levels $(5.0 \times 10^{-5} \text{ a}^{-1})$. Non-carcinogenic risks were lower than the allowance levels (10⁻⁶ a⁻¹). Children had greater health risks than adults. The health risks of metals through the drinking pathway were 2-3 orders of magnitude higher than the values caused by the dermal contact pathway. For the sake of drinking water safety, the well water, underground river water, and spring water should be properly treated and the concentration of Cr in groundwater should be controlled before drinking.

Key words: groundwater; water quality analysis; Nemerow index; health risk assessment; area of Xiangshui, Chongzuo

崇左市位于广西壮族自治区西南部,地貌以喀斯特岩溶地貌为主体,多出露石炭系、二叠系和三叠系地层,碳酸盐岩地层面积为433.04 km²,占研究区面积的91%,岩性以灰白色-灰色中厚-厚层灰岩为主,褶皱和断层发育,地下河和岩溶泉发育,防污性能极其脆弱.该市地处"南宁—新加坡经济走廊"的节点,经济以农业为主,甘蔗为主要的经济作物,矿产资源丰富.随着工业化和城市化的快速发展,甘蔗生长期间肥料使用、农药喷洒、蔗糖加工及矿山开采等人类活动增加,造成大量有害物

质沉积地表并通过各种途径进入到含水层,极易污染含水层中的地下水,地下水资源正面临着严重的威胁^[1~3].水质评价是研究人类水环境质量变化规律,评价人类水环境质量水平,并对水环境要素或区域水环境性质的优劣进行定量描述的科学^[4],对

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该地区经济的可持续发展有着重要意义.金属元素 具有稳定性、生物积累性和不可降解性等特点,排 放到环境后不断迁移、转化进入并累积于空气、土 壤或水体中.金属元素是地下水中常见的污染因 子,它在地下水中的迁移转化是一个复杂的过程, 以机械迁移、物理化学迁移和生物迁移等方式迁 移,其在地下水中通过吸附、沉降、释放和生物吸 收等过程进行物理、化学及生物反应,涉及溶解作 用、沉淀作用、吸附作用和氧化还原作用等转化机 制,并能通过食物链在生物体内富集,危害身体健 康.地下水一定剂量的金属元素经长时间暴露会对 人体产生致癌和非致癌等健康风险^[5~8].

目前,关于水质评价方法的研究集中在单因子 指数法、内梅罗综合指数法、模糊综合法和水质生 物综合评价法等方法的优缺点、改进和应用等方 面[9~15],健康风险评价多见于电镀厂、水库、饮用 水源、污灌区等的健康风险评价[16~20]. 而将水质 评价及健康风险评价相结合对地下水质量进行评价 的研究较少. 将健康风险评价与水质评价相结合, 能更全面地掌握地下水环境质量,了解地下水安全 状况,有助于加强地下水的风险管理,制定和实施 相应的污染物控制策略. 本文采用内梅罗综合指数 法和健康风险评价相结合的方法, 以崇左响水地区 井水、泉水和地下河水3种类型地下水中常规指标 及金属元素的质量浓度测试结果为依据,采用内梅 罗综合指数法分析了3种不同类型地下水的质量级 别,采用健康风险评价模型对地下水中金属元素通 过饮水和皮肤接触途径产生的健康风险进行了评 价,以期为研究区地下水的环境质量提供更全面的 参考资料,并为地下水金属元素污染水质风险管

理,水资源保护及人类健康保障提供科学依据.

40 卷

1 材料与方法

1.1 研究区概况

本研究选取崇左响水地区(22°10′~22°20′N, 107°00′~107°45′E)为研究区域. 研究区位于崇左 市的中西部,属亚热带季风气候区,年平均气温 20.8℃~22.4℃,年无霜期340 d,年平均日照时数 1600 h, 年平均降雨量1199 mm, 6~9 月为丰水 期,5、10、11月为平水期,12月~次年4月为枯水 期. 研究区河流属于左江流域, 地表河流主要为左 江干流,黑水河,以及由岩溶水调节的季节性小 河,流长81.6 km. 研究区地势南北部较高,中东部 较低, 由西北向东部倾斜, 海拔高度 150~500 m, 中部被左江及支流切割,形成丘陵平原[21~23].研 究区地层多以石炭系、二叠系和三叠系为地质基 层,岩性以灰白色-灰色中厚-厚层灰岩为主,页岩、 砂岩次之, 第四系酸性赤红壤土层为地表盖层, 在 研究区西北角分布少量白云岩, 二叠系上统合山组 地层(P3h)为紫红色薄层状铁质泥岩、豆状铁铝岩. 研究区岩溶发育强烈, 地下水水量丰富, 主要接受 大气降水补给, 地下水径流条件好, 地下水以地下 河出口和岩溶泉的形式分别由南、北、西往中部的 左江排泄. 研究区矿产资源较为丰富, 分布有铁 矿、铅锌矿、煤矿、锰矿、制糖和造纸等生产企业, 经济以农业为主,甘蔗为主要的经济作物,肥料使 用、农药喷洒、蔗糖加工及矿山开采对地下水造成 污染, 威胁生态环境, 危害人体健康[24].

1.2 样品采集与测试

如图 1 所示, 本研究于 2016 年 7 月在研究区采

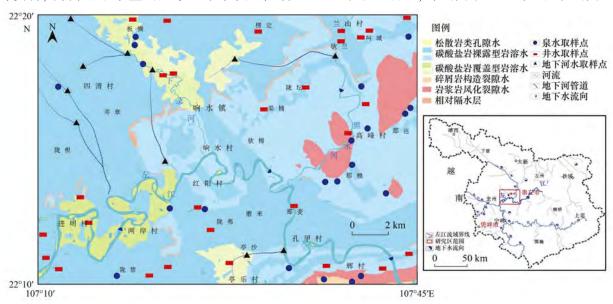


图 1 研究区地理位置和采样点分布示意

Fig. 1 Location of the study area and distribution of sampling sites

集60个样点的地下水样品,其中,井水样品28个,泉水样品21个,地下河水样品11个,研究区地理位置和采样点分布见图1.2016年7月为丰水期,降雨量为749.5 mm,占全年降雨量的63.5%.另于枯水期2016年3月、平水期5和11月分别在研究区进行采样,受人类活动影响,K⁺、Na⁺、Cl⁻和NO₃⁻平均质量浓度表现为丰水期>平水期>枯水期,因平水期径流较小,大部分金属元素在平水期较丰水期大,其余指标无明显的季节性变化。由于数据量过大,同时考虑到研究区域的自然气候状况,认为丰水期的水质状况更具有区域整体代表性,因此采用丰水期2016年7月的数据进行水质分析及健康风险评价.

样品测试采用仪器现场测定和室内测试相结合 的研究方法. 现场测定使用 Multi340i 便携式水质 多参数分析仪(WTW)测定各水点 pH 值,测试精度 为 0.01 个 pH 单位. 用聚乙烯瓶采集 1.5L 地下水 样品,取样前先用去离子水清洗聚乙烯瓶3次,再 用所采水样润洗3次,采集后立即用0.45 µm 水系 微孔滤膜过滤,滤液装入聚乙烯瓶中. 用于测定的 阳离子(Na+、NH, 和 Fe)水样立即加入超纯硝酸 酸化使其 pH < 2, 带回实验室后放置于 4℃冰箱中 密封保存;用于测定阴离子(Cl-、NO, 、SO4-和 NO;)的水样直接于 4℃ 冰箱中密封保存; 用于 测定金属元素(Zn、Fe、Al、Cr、Mn、Cu、Pb、Cd、 Hg 和 As)的水样加入 HNO₃(1:1)酸化, HNO₃含量 为1%,带回实验室置于4℃冰箱中避光保存.采用 滴定管测定高锰酸盐指数,检测依据为 DZ/T 0064.88-93.0; 采用全谱直读等离子体光谱仪(IRIS Intrepid Ⅱ XSP, Thermo Electron)测定水样中阳离 子的质量浓度, 检测依据为 GB/T 5750.6-2006; 采 用离子色谱仪(ICS-1100, Dionex)测定水样阴离子

的质量浓度,检测依据为 DZ/T 0064.51-93;采用电感耦合等离子体质谱仪(iCAP Q, Thermo Fisher)测定水样中金属元素质量浓度,检测依据为 GB/T 5750.6-2006. 经检测,地下水样品中各指标测定值的相对标准偏差(RSD)均小于 5.0%,加标回收率为 80%~120%.

1.3 内梅罗综合指数法

内梅罗综合指数法是一种采用加附注,兼顾极值或突出最大值的计权型多因子环境质量评价的方法. 要求选择参评项目不少于标准规定的监测项目,不包括细菌学指标,能反映本地区主要水质问题. 首先,进行各单项组分评价,参与评分的因子标准依据《地下水质量标准》(GB/T 14848-2017)规定的 工类标准限值,划分单项组分所属的质量级别,根据地下水单项组分评分(表1)确定单项组分的平均分值 F_i ; 其次,根据式(1)和式(2),选用内梅罗综合指数计算综合评价分值 F; 最后,根据综合评价分值 F 和地下水质量分级(表2)确定地下水质量级别[25-27].

表 1 地下水单项组分评分

Tab	le 1 Grade	s of single co	omponents in	ı groundwat	er
水质类别	(A)	II V	d II	IV	By.
\boldsymbol{F}_i	0	10 9	3	6_	10

$$F = \sqrt{\frac{F_{\text{max}}^2 + \overline{F}^2}{2}} \tag{1}$$

$$\overline{F} = \frac{1}{n} \sum_{i=1}^{n} F_i \tag{2}$$

式中,F 为综合评价分值; \overline{F} 为各单项组分评分值 F_i 的平均值; F_i 为各单项组分评分值; F_{max} 为单项组分评分值 F_i 中的最大值;n 为项数.

表 2 地下水质量分级

Table 2 Grades of groundwater quality

			- Broantan area quarrey		
级别	优良	良好	较好	较差	极差
\overline{F}	< 0.80	0. 80 ~ 2. 50	2. 50 ~ 4. 25	4. 25 ~ 7. 20	>7. 20

1.4 健康风险评价

健康风险评价是通过估算有害因子对人体不良影响发生的概率来评价暴露于该因子的个体健康受到影响的风险. 水体中金属元素通过饮水和皮肤接触 2 种途径暴露^[28]. 地下水中金属元素对人体健康产生危害的风险类型分为化学致癌性金属所致健康危害的风险和非致癌性金属所致健康危害的风险和非致癌性金属所致健康危害的风险. 根据世界卫生组织 WHO 和国际癌症研究机构IARC 对所检测项目化学物质致癌性的可靠性程度全面分析评价,将研究测定的 10 种金属元素分为

化学致癌性金属元素 As、Cd、Cr 和化学非致癌性 金属元素 Al、Cu、Fe、Hg、Mn、Pb、Zn. 化学致癌 性与非致癌性金属元素的健康风险评价模型不同, 模型及参数 PC、SF 和 RfD 值参照文献[29~33].

2 结果与讨论

2.1 水质分析

首先,对地下水各水质指标数值的分布类型进行正态性检验,少部分指标 pH 值、TDS、总硬度(CaCO₃)和 Cr 质量浓度符合正态分布,多数指标符

合对数正态分布.不同类型地下水水质常规指标统计见表 3. 依据《地下水质量标准》(GB/T 14848-2017)规定的Ⅲ类标准限值,井水、泉水和地下河水中 NO₃ 部分水点质量浓度超过Ⅲ类标准限值,超标率为 60.71%、52.38%和 36.36%,最大值分别是标准限值的 4.84、4.21 和 2.01 倍;泉水中 F⁻、NH₄ 和高锰酸盐指数在部分水点出现超标现象,超标率为 9.52%、4.76% 和 4.76%,最大值分别是标准限

值的 1.5、4.68 和 1.17 倍;地下河水 NO_2^- 部分水点质量浓度超过 \mathbb{II} 类标准限值,超标率为 18.18%,最大值分别是标准限值的 4.5 倍.井水、泉水和地下河水中 NO_2^- 、 NH_4^+ 、 F^- 和高锰酸盐指数变异系数均超过 100%,泉水中 SO_4^{2-} 、 Na^+ 变异系数超过 100%,江河水中 NO_3^- 变异系数超过 100%,说明 NO_2^- 、 NH_4^+ 、 F^- 和高锰酸盐指数在井水、泉水和地下河水各采样点均表现出较大幅度的变化.

表 3 不同类型地下水水质常规指标统计/mg·L-1

Table 3	Romiler water	quality	indoves in	different	tymog of	groundwater/mg	.T -1
rabie 5	negular water	quantv	indexes in	amerent	types of	groundwater/ mg	· L

		1 an	de 3 - Regi	mai water c	Įuality index	es in umei	em types o	n groundwai	er/ mg·r			
地下 水类型	统计量	pH 值	SO ₄ ²⁻	Cl -	F -	NO ₃	NO ₂	NH ₄ ⁺	Na +	高锰酸盐 指数	总硬度 (CaCO ₃)	TDS
	平均值	7. 20	12. 22	15. 34	0.04	35. 85	0.06	0. 01	3. 78	0.46	311.74	345. 43
	最大值	7. 63	39.42	53.84	0. 19	96. 88	0.71	0.08	13.44	2. 98	440. 85	530. 26
	最小值	6. 77	1.45	2. 27	nd^{1}	7.46	nd	nd	0.34	nd	197. 53	208. 33
井水	中值	7. 18	10.43	9. 57	nd	28. 05	$^{\mathrm{nd}}$	nd	2. 22	nd	311.94	337. 04
71/10	标准差	0. 22	9. 26	13.45	0.06	25. 58	0.18	0.02	3.72	0.68	68. 11	86. 63
	变异系数/%	3.00	75. 79	87. 68	166. 80	71. 34	274. 09	176. 99	98. 47	148. 43	21. 85	25. 08
	超标率/%	0.00	0.00	0.00	0.00	60.71	0.00	0.00	0.00	0.00	0.00	0.00
	标准限值2)	6.5 ~ 8.5	250.00	250.00	1.00	20.00	1.00	0.50	200.00	3.00	450. 00	1 000. 00
	平均值	7. 14	16. 19	15.81	0.20	33. 67	0.01	0. 13	3. 71	0.65	302. 10	335. 18
	最大值	7. 91	82. 84	49. 52	1.50	84. 13	0.07	2.34	18. 11	3. 51	422. 23	471. 22
	最小值	9. 74	4. 44	1.51	nd	nd	nd	nd	0.41	nd	8. 26	154. 90
泉水	标准差	0. 27	16. 44	14. 33	0.42	27. 81	0.02	0.51	4. 38	0. 95	87. 67	102. 25
ACAL	中值	7. 07	13. 57	11.55	0.07	26. 50	nd	nd	1. 79	nd	299. 57	353. 83
/	变异系数/%	3. 83	101. 57	90. 61	208. 97	82. 59	238. 32	397. 65	118. 25	147. 27	29. 02	30. 50
9	超标率/%	0.00	0.00	0.00	9, 52	52. 38	0.00	4.76	0.00	4. 76	0.00	0.00
	标准限值2)	6.5 ~ 8.5	250. 00	250.00	1.00	20.00	1. 00	0.50	200.00	3. 00	450. 00	1 000. 00
1	平均值	7. 19	7. 44	8. 15	0.05	17. 24	0. 51	0.01	1. 49	0.56	254. 27	265.66
10	最大值	7. 52	12.00	13.06	0. 14	40. 22	4. 50	0.04	3. 73	1. 63	306. 08	310. 98
19	最小值	6. 76	3. 95	2. 55	/ Ind	4. 30	nd	nd	0.72	nd	153. 19	169. 97
地下	中值	7. 21	7.73	8. 39	0. 03	11.66	nd	nd	1. 22	nd	259. 93	280. 15
河水	标准差	0. 23	2.47	3.68	0.05	11. 94	1. 36	0.01	0.82	0.74	46. 79	44. 97
41	变异系数/%	3. 17	33. 22	45. 10	112. 56	69. 22	267. 52	185.40	54. 85	132. 79	18.40	16. 93
	超标率/%	0.00	0.00	0.00	0.00	36. 36	18. 18	0.00	0.00	0.00	0.00	0.00
	标准限值2)	6.5 ~ 8.5	250.00	250.00	1.00	20.00	1.00	0.50	200.00	3.00	450.00	1 000. 00

1) nd:未检出; 2) 地下水质量标准Ⅲ类标准(GB/T 14848-2017), 下同

不同类型地下水金属元素质量浓度统计见表 4. 依据《地下水质量标准》(GB/T 14848-2017), 井水中 Zn 和 Hg 部分水点质量浓度超过Ⅲ类标准限值,超标率为 7.14% 和 3.57%,最大值分别是标准限值的 1.58 倍和 1.52 倍,泉水中 Fe、Mn和 Al部分水点质量浓度超过Ⅲ类标准限值,超标率为 9.52%、4.76% 和 4.76%,最大值分别是标准限值的 5.90、9.28 和 1.25 倍,地下河水中 Hg 部分水点质量浓度超过Ⅲ类标准限值,超标率为 4.76%,最大值是标准限值的 21.10 倍. 井水、泉水和地下河水中 Fe、Mn、Al、Cu、Zn、Hg 和 As 变异系数超过 100%,地下河水中 Cd 变异系数超过 100%,金属测定方法本身的变异系数在 10% 以内,说明 Fe、Mn、

Al、Cu、Zn、Hg和As质量浓度在井水、泉水和地下河水各采样点均表现出较大幅度的变化. Fe、Mn和Al质量浓度较高的水点主要分布在研究区红阳村附近,该地出露二叠系上统合山组(P₃h)紫红色薄层状铁质泥岩,含较多的铁矿物,铁矿多为露天开采,造成该处水点中Fe及其伴生矿物元素Mn、Al质量浓度较高;Cu、Zn、Hg和As质量浓度较高的水点主要分布在研究区进明村、红阳村、高峰村、孔甲村、辉村等村庄和农田密集区,畜禽饲料及粪便中含Cu、Zn、Hg和As等金属元素且农药、化肥的连续施用会增加地下水中Cu、Zn、Hg和As的质量浓度,导致Fe、Mn、Al、Cu、Zn、Hg和As在各采样点表现出较大幅度的变化,变异系数超过100%.

表 4	不同為	类型地下	水金属元素	质量浓度统计	∵/µg•	L-1	

			Fable 4 Met	tal concentra	tions in diff ϵ	erent types of	groundwater	∕µg•L ⁻ ¹			
地下 水类型	统计量	Fe	Mn	Al	Cu	Zn	Pb	Cr	Cd	As	Hg
	平均值	5. 47	1. 97	4. 76	1. 29	175. 84	0. 67	4. 34	0. 23	0. 10	0. 11
	最大值	51.00	10.00	25. 30	6.03	1583.00	2.74	7.88	0.51	0.58	1.52
	最小值	nd	nd	nd	nd	nd	nd	2. 24	nd	nd	nd
井水	中值	nd	0.89	2. 58	0.61	13. 85	0.64	4. 11	0. 24	nd	nd
7171	标准差	10.70	2.72	6. 65	1. 75	393. 17	0.59	1.30	0. 15	0. 17	0.34
	变异系数/%	195. 75	137. 98	139. 82	135. 57	223.60	88. 59	30.03	64. 37	163.48	320. 80
	超标率/%	0.00	0.00	0.00	0.00	7. 14	0.00	0.00	0.00	0.00	3. 57
	标准限值	300.00	100.00	200.00	1 000.00	1 000. 00	10.00	50.00	5. 00	10.00	1.00
	平均值	105. 45	49. 14	29. 47	0.41	45. 46	0.70	3. 35	0. 20	0.77	0.06
	最大值	1770.00	928.00	250.00	3. 76	919.00	2. 26	5. 12	0.37	7. 92	0.67
	最小值	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
泉水	中值	0.00	0.40	1. 79	0.11	0.00	0.51	3.58	0.18	0.39	0.00
76/10	标准差	391. 22	201.70	64. 56	0.84	200. 20	0.50	1. 21	0. 11	1.69	0. 17
	变异系数/%	371.01	410. 44	219.05	204. 96	440. 36	71. 23	35. 95	57.00	219. 44	271.97
	超标率/%	9. 52	4. 76	4. 76	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	标准限值	300.00	100.00	200.00	1 000.00	1 000.00	10.00	50.00	5. 00	10.00	1.00
	平均值	2. 63	6. 97	15. 76	0. 17	37. 43	0. 43	3. 65	0. 14	0. 37	1. 95
	最大值	15.00	29. 20	79. 80	0.66	328.00	1. 19	6.86	0.38	1. 67	21. 10
	最小值	nd	nd	1.00	nd	nd	nd	1. 97	nd	nd	nd
地下	中值	nd	1.50	7. 21	d nd	nd	0.50	2. 92	nd	nd /	nd
河水	标准差	5. 03	12. 08	23. 26	0. 26	97. 47	0.41	1.66	0. 17	0.56	6. 35
1,1/1/	变异系数/%	191. 27	173. 28	147. 61	153. 75	260.38	97. 02	45. 38	118. 22	151.60	325. 42
	超标率/%	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	4. 76
_	标准限值	300.00	100.00	200.00	1 000.00	1 000. 00	10.00	50.00	5. 00	10.00	1.00

从地下水水质常规指标平均质量浓度的分布特征(图 2)和金属元素平均质量浓度的空间分布特征(图 3)可知, 井水中 pH 值、TDS、总硬度(CaCO₃)、NO₃ $^-$ 、Na $^+$ 、Zn、Cu、Cr 和 Cd 平均质量浓度最高, 泉水中高锰酸盐指数、SO₄ $^-$ 、Cl $^-$ 、F $^-$ 、NH₄ $^+$ 、Fe 和 Mn 平均质量浓度最高,地下河水中 NO₂ $^-$ 和 Hg 平均质量浓度最高。在井水、泉水和地下河水中 Zn、Fe、Al、Cr 和 Mn 平均质量浓度均高于 Cu、Pb、Cd、Hg 和 As. 按井水、泉水和地下河水中金属元素平均质量浓度堆积总浓度大小顺序依次为泉水

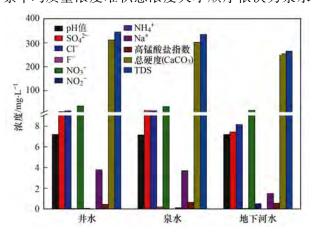


Fig. 2 Distribution characteristics of regular water quality indexes in different types of groundwater

水质常规指标在不同类型地下水的分布特征

(235. 02 μ g·L⁻¹) > 井水(194. 7 μ g·L⁻¹) > 地下河 水(69. 50 μ g·L⁻¹). 对不同类型地下水中水质参数 进行 Kruskal-Wallis 检验, TDS 和 SO₄² 质量浓度均 为差异性显著, Cu、Zn 和 As 质量浓度均为差异性 极显著, 其他指标差异不显著.

研究区地下水水质指标具有较明显的空间变化规律. 研究区地下水分别由南、北、西这 3 个方向往中部的左江排泄,位于研究区西部的两岸村等地因其上游分布的糖厂和造纸厂,在造纸过程中产生的大量 Hg 使得该处 Hg 质量浓度高于其他区域;位于研究区中部的红阳村 Fe、Mn 和 Al 质量浓度最

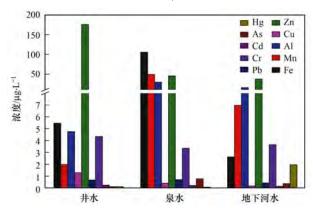


图 3 金属元素平均质量浓度在不同类型地下水的分布特征

Fig. 3 Distribution characteristics of average concentrations of metals in different types of groundwater

高,因为红阳村距离铁矿最近,其与当地的自然地质条件和矿产开采有密切关系;位于红阳村下游的亭乐村 Fe、Mn 和 Al 质量浓度较高与接受上游红阳村地下水补给有关;位于研究区东北部的兰山村 Zn 质量浓度偏高,与接受北部铅锌矿在开采过程中排放的含 Zn 废水补给有关;位于研究区进明村、红阳村、高峰村、孔甲村、辉村等村庄和农田密集区 NO_3^- 、 NO_2^- 、 NH_4^+ 和高锰酸盐指数质量浓度偏高,与种植农作物期间化肥的连续施、大量喷洒农药和畜禽饲料使用及粪便排放有关.

地下水单项组分评分结果见表 5,表 5 中仅列出了水质类别超过 \mathbb{II} 类(\mathbb{IV} 和 \mathbb{V}) 的组分,井水中各水点水质类别超过 \mathbb{II} 类组分有 \mathbb{NO}_3^- 、 \mathbb{Z} n 和 \mathbb{Hg} ,泉水中水质类别超过 \mathbb{II} 类组分有 \mathbb{NO}_3^- 、 \mathbb{F}^- 、 \mathbb{NH}_4^+ 、高锰酸盐指数、 \mathbb{F} e、 \mathbb{M} n 和 \mathbb{A} l,地下河水中水质类别超过 \mathbb{II} 类组分有 \mathbb{NO}_3^- 、 \mathbb{NO}_2^- 和 \mathbb{Hg} . 研究区大量种植甘蔗,农业施肥、大面积甘蔗秸秆燃烧、人畜排泄

物、土壤有机氮和 NH⁺ 硝化作用造成 NO⁻、大面 积面源污染;区内磷肥厂、化工厂、冶炼厂使用含 氟矿石等工业活动和生产生活过程中煤炭燃烧排 出大量含氟粉尘、废气造成 F-质量浓度较高; NH; 与枯树等植物腐烂有关; 高锰酸盐指数与甘 蔗生长期间大量喷洒农药产生的有机污染物及还 原性物质有关;结合研究区地质条件,二叠系上 统合山组(P,h)紫红色薄层状铁质泥岩含较多的 铁矿物,如:赤铁矿(Fe,O,)、褐铁矿(2Fe,O,* 3H,O)和针铁矿等,铁矿多为露天开采,造成Fe 及其伴生矿物元素 Mn、Al 质量浓度较高;研究区 北部分布有铅锌矿, 铅锌矿开采过程中含 Zn 废水 排放及矿渣堆存增加了地下水中 Zn 的质量浓度: 研究区两岸村上游分布的糖厂和造纸厂, 造纸过 程中对造纸白泥进行资源化处理的同时产生了大 量 Hg, Hg 源源不断进入地下水,造成地下水中 Hg 质量浓度较高.

表 5 地下水单项组分评分结果

Table 5	Evolution	roculta	for	ainala	aamnananta	in	groundwater
rabie 3	Evaluation	resuits	IOL	single	components	ın	groundwater

1.1 - 1. M. mit -											
地下水类型	项目	NO_3^-	F -	NO_2^-	$\mathrm{NH_4}^+$	TFe	高锰酸盐指数	Mn	Al	Zn	Hg
井水 样	品数	17	0	0	0	0	0 /	0	0	2	10
超板	示率/%	60.71	0.00	0.00	0.00	0.00	0.00	0.00	0.00	7. 14	3. 57
泉水	品数	11	2	0/	D.M.	2	11/1	10	11	0	(0//
超标	示率/%	52. 38	9.52	0.00	4. 76	9. 52	4. 76	4. 76	4. 76	0.00	0.00
地下河水	品数	4	0	2	0	0	0/1	0	0	0	7 6
超板	示率/%	36. 36	0.00	18. 18	0.00	0.00	0.00	0.00	0.00	0.00	4. 76

根据地下水质量级别评价结果可知(表6),井水、泉水和地下河水的综合评价分值分别为7.10、7.10和4.26,水质级别均为较差级别.井水和地下河水各采样点的水质级别有较好和较差2种,泉水有较好、较差和极差3种.地下河水各采样点水质级别为良好的样品数百分比最大(36.36%),井水(35.71%)次之,泉水(33.33%)最小,水质级别为较差的样品数均占到了60%以上.内梅罗综合指数法根据相应的分级标准确定水质类型,在各组分水质分级明显、超标组分较多或者没有超标组分的情况下可以较合理地反映水质状况.它能够极突出最大污染因素,即使参评项目中只有一项组分值偏高.地

下水中大多数组分超标率较低,但存在超标率较高的组分,如NO3⁻ 在井水、泉水和地下河水中的超标率分别为60.71%、52.38%和36.36%,并且各超标组分分布在不同的水点中,所以综合评价分值较高的水点数较多,水质级别为较差的样品数均占到了60%以上.根据《地下水质量标准》(GB/T14848-2017)要求,地下水质量为良好级别的井水、泉水和地下河水的水点地下水化学组分含量为天然背景含量,适用于各种用途;为较差级别的井水、泉水和地下河水的水点地下水化学组分含量较高,以农业和工业用水质量要求以及一定水平的人体健康风险为依据适用于农业和部分工业用水,适当处理后可作为生活饮用水.

表 6 地下水质量级别评价结果

Table 6 Evaluation results for groundwater quality grades

地下水类型	F	项目	优良	良好	较好	较差	极差	总数
井水	7. 10	样品数	0	10	0	18	0	28
<i></i>	7. 10	样品数百分比/%	0.00	35.71	0.00	64. 29	0.00	100.00
泉水 7.10		样品数	0	7	0	13	1	21
永小 7.10	样品数百分比/%	0.00	33. 33	0.00	61. 90	4. 76	100.00	
地下河水	4. 26	样品数	0	4	0	7	0	11
地下河水	4. 20	样品数百分比/%	0.00	36. 36	0.00	63. 64	0.00	100.00

2.2 相关分析

根据地下水金属元素与其它组分之间的相关系数可知(表7),pH值与Cu和As之间呈极显著正相关,表明地下水的pH值影响着Cu和As质量浓度的分布,Cu和As的形成和迁移富集与酸碱度有密切关系,随着pH值的增高,土壤胶体上正电荷减少,对As的吸附量降低,相应地可溶性含量增高,致使地下水中As质量浓度也增高;总硬度(CaCO₃)、TDS与Cr之间呈显著正相关,总硬度(CaCO₃)、TDS与Mn、Al、Fe呈显著或极显著负相关,说明高硬度、高矿化度有利于Cr的富集,不利于Fe、Mn和Al的富集,可能与高矿化度增加了水对离子态含铬化合物的溶解度有关;高锰酸盐指

数、NH₄⁺、F⁻与 As 之间呈显著或极显著正相关,由于高锰酸盐指数、NH₄⁺和 F⁻代表地下水处于还原环境,说明还原环境有利于 As 的富集,因为氧化还原作用制约着 As 在环境中存在的形式和迁移能力,随着电位的降低,还原性的增强,元素被还原成比较容易溶解的低价态形式,可溶性含量增高,地下水的 As 质量浓度增高;Na⁺与金属元素之间无显著性相关关系,NO₃⁻除与部分金属元素呈显著负相关外,与其它金属之间无显著性相关关系,说明 Na⁺和 NO₃⁻不是影响地下水金属元素质量浓度分布的主要影响因子;Zn 和 Hg 与其它组分的相关性均不明显,说明 Zn 和 Hg 受其它组分的影响不明显.

表 7 地下水金属元素与其它组分之间的相关系数1)

Table 7 Correlation matrix between metals and other components in groundwater	Table 7	Correlation	matrix	between	metals	and	other	components	in	groundwater
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							0			
	Fe	Mn	Al	Cu	Zn	Pb	Cr	Cd	As	Hg
pH 值	-0.090	0.090	0. 156	0. 408 **	0. 147	0. 166	-0.252	0. 227	0. 349 **	-0.177
SO ₄ ^{2 -}	-0.373 **	-0.103	-0.303*	0.045	0. 111	0. 292 *	-0.196	0. 206	0. 323 *	-0.075
Cl -	-0.066	-0.138	-0.105	0. 158	0.006	0. 284 *	0.013	0. 210	0. 205	-0.069
F -	-0.097	0. 210	0. 134	0. 355 **	-0.183	0. 200	-0.110	0. 417 **	0. 798 **	0.073
NO_3^-	-0.209	-0.414**	-0. 265 *	-0.028	0. 177	0. 091	0. 154	-0.083	-0. 289 *	-0. 225
NO_2^-	-0.011	0.046	0. 022	0.039	-0.023	-0.047	0. 300 *	0. 023	0. 051	0.041
NH ₄ ⁺	0. 293 *	0. 257 */	0. 116	0. 259 *	-0.186	0. 263 *	-0.106	0. 352 **	0. 392 **	0. 208
Na +	-0.077	-0.134	-0.097	0. 126	0.025	0.094	0. 124	0.037	0.054	-0.108
高锰酸盐指数	0. 263 *	0. 358 **	0. 386 **	0. 475 **	-0.136	0. 223	0.094	0. 433 **	0. 564 **	0.012
总硬度(CaCO ₃)	/-0. 298 *	-0.385 **	-0.463**	-0.128	0. 134	0. 059	0. 450 **	-0.104	-0.211	-0.091
TDS	-0. 270 *	-0.362 **	-0. 408 **	-0.047	0. 149	0. 101	0. 433 **	-0.056	-0.173	- 0. 102
1			E/ //	DF 10		1 9 1			•	10

1)**表示在置信度(双测)为0.01时,相关性是显著的;*表示在置信度(双测)为0.05时,相关性是显著的,下同

采用 Spearman 相关系数分析地下水金属元素之间的相关关系,根据地下水金属元素之间的相关系数可知(表 8), Zn、Pb、Cd 和 Cu 之间存在显著或极显著的正相关关系,显示地下水中 Zn、Pb、Cd和 Cu 可能经过相似的环境地球化学作用。曾昭华[34]的研究结果表明 Zn、Pb、Cd和 Cu 均为亲铜元素,Cd 的地球化学行为与 Zn 相似,在酸性介质和氧化条件下,Zn、Pb、Cd和 Cu 多呈稳定的离子状态而有利于迁移富集,在碱性介质和还原环境中

趋于沉淀; Fe、Al 和 Mn 之间存在显著或极显著的正相关关系,显示地下水中 Fe、Al 和 Mn 经过相似的环境地球化学作用,来源相似.曾昭华[35]的研究结果表明 Fe 和 Mn 易在还原环境中形成,结合研究区地质条件, Al、Mn 作为 Fe 的伴生矿物和 Fe 均主要来源于二叠系上统合山组(P₃h)紫红色薄层状铁质泥岩地层的铁矿开采; Hg 和 As 之间呈显著正相关,显示 Hg 和 As 可能具有相似的空间变化规律,研究区进明村、红阳村、高峰村、孔甲村和辉村等

表8 地下水金属元素之间的相关系数

Table 8 Correlation matrix of metals in groundwater

	Fe	Mn	Al	Cu	Zn	Pb	Cr	Cd	As	Hg
Fe	1	0. 318 *	0. 519 **	0. 397 **	0.014	-0.116	0.068	-0.067	-0.126	-0. 081
Mn		1	0. 525 **	0. 150	0.002	0. 219	-0.095	0. 342 **	0. 303 *	0. 220
Al			1	0. 259 *	-0.093	0.005	-0.198	0.034	0.095	0.037
Cu				1	0. 356 **	0. 356 **	0. 107	0. 292 *	0. 207	-0.060
Zn					1	0. 259 *	0.014	0.082	-0.347 **	-0.144
Pb						1	-0.061	0. 762 **	0. 252	0.009
Cr							1	0.079	-0.249	-0.072
Cd								1	0.412 **	0.055
As									1	0. 285 *
Hg										1

村庄和农田密集区,畜禽饲料及粪便中含 Cu、Zn、Hg 和 As 等金属元素且农药、化肥的连续施用会增加地下水中 Hg 和 As 的质量浓度; Zn 与 As 之间呈极显著负相关, Cr 与其它金属元素间无显著性相关关系,显示 Zn 与 As、Cr 与其它金属无相似的空间变化规律.

2.3 健康风险评价

依据健康风险评价模型、模型参数和金属元素 质量浓度数据,计算出地下水金属元素经饮水途径 (表9)和皮肤接触途径(表10)所引起的个人平均 年健康风险.

由地下水金属元素经饮水和皮肤接触途径引起 的个人平均年健康风险结果可知, 研究区地下水金 属元素致癌风险偏高,总致癌风险数量级为10-9~ 10-5, 总致癌风险大小顺序为井水 > 地下河水 > 泉 水. 成人和儿童的总致癌风险分别在 7.40×10⁻⁵~ 9.50×10^{-5} a^{-1} 和 $8.05 \times 10^{-5} \sim 1.03 \times 10^{-4}$ a^{-1} 之 间,儿童总致癌风险均高于成人. 井水、泉水和地 下河水的个人平均年致癌风险均高于国际辐射防护 委员会(ICRP)推荐的最大可接受风险水平 5.0 × 10⁻⁵ a⁻¹, 致癌风险均来源于 Cr, Cr 个人平均年致 癌总风险值高于 5.0×10^{-5} a⁻¹, 而 As 和 Cd 低于 5.0×10⁻⁵ a⁻¹. As、Cd 和 Cr 质量浓度均满足Ⅲ类 水质标准, 但具有较高的致癌风险, 是因为致癌风 险不仅与 As、Cd 和 Cr 质量浓度有关, 还与致癌强 度系数、人体日平均饮水量、暴露频率、暴露持续 时间、人均体重、平均暴露时间和人类平均寿命有 关. 经饮水和皮肤接触途径引起的个人平均年致癌 风险分别在 $8.00 \times 10^{-8} \sim 1.02 \times 10^{-4} \ a^{-1}$ 和 $1.28 \times$ 10⁻⁹~9.66×10⁻⁷ a⁻¹之间. 井水、泉水和地下河水 的总致癌风险来源物质顺序为 Cr > Cd > As, 其中, 经饮水途径引起的个人平均年致癌风险的来源物质 顺序均为 Cr > Cd > As, 井水经皮肤接触途径引起 的致癌风险来源物质顺序为 Cr > Cd > As, 泉水和

地下河水为 Cr > As > Cd.

研究区 3 种类型地下水的总非致癌风险数量级为 $10^{-15} \sim 10^{-9}$,总非致癌风险大小顺序为地下河水 > 泉水 > 井水. 成人和儿童的总非致癌风险在8.39 × $10^{-10} \sim 3.82 \times 10^{-9} a^{-1}$ 和 9.03 × $10^{-10} \sim 4.15 \times 10^{-9} a^{-1}$ 之间,均小于可以接受的健康风险水平 $10^{-6} a^{-1}$,儿童的总非致癌风险均大于成人. 经饮水途径和皮肤接触途径引起的个人平均年非致癌风险分别在 2.24 × $10^{-12} \sim 3.72 \times 10^{-9} a^{-1}$ 和 7.72 × $10^{-15} \sim 3.18 \times 10^{-11} a^{-1}$ 之间. 井水的总非致癌风险来源物质顺序为 Zn > Pb > Hg > Mn > Al > Cu > Fe,泉水为 $Mn > Pb > Fe > Al > Hg > Zn > Cu,地下河水为 <math>Hg > Pb > Mn > Zn > Al > Fe > Cu,7 种非致癌性金属元素对人体健康产生危害的个人平均年健康风险水平集中在 <math>10^{-15} \sim 10^{-9} a^{-1}$,所引起的健康风险较小,不会对暴露人群构成明显危害.

研究区地下水金属元素所引起的健康总风险偏 高,总风险大小顺序为井水>地下河水>泉水,与 3 种类型地下水中 Cr 质量浓度的大小顺序一致. 从 金属元素的暴露途径来说, 经饮水途径引起的健康 风险比和皮肤接触途径高2~3个数量级,与余葱 葱等[16]对电镀厂周边地表水中重金属分布特征及 健康风险评价研究结果一致,表明饮水途径是金属 元素的主要暴露途径. 成人和儿童的总风险分别在 $7.40 \times 10^{-5} \sim 9.50 \times 10^{-5} \ a^{-1}$ 和 $8.05 \times 10^{-5} \sim 1.03$ ×10⁻⁴a⁻¹之间,均高于5.0×10⁻⁵a⁻¹,3种类型地 下水儿童总风险均大于成人,说明较成人而言,儿 童是更加敏感的风险受体, 受到金属的危害更严 重,其中,儿童经饮水途径引起的健康总风险大于 成人, 而经皮肤接触途径引起的健康总风险小于成 人, 这与研究选取的模型参数皮肤接触面积 SA, 人 均体重 BW, 暴露频率 ET 有关, 此结果与张清华 等[36]对柳江流域饮用水源地重金属污染与健康风 险评价研究结果相近,因此应对儿童的饮水安全进

表 9 不同类型地下水金属元素经饮水途径引起的个人平均年健康风险/a-1

Table 9 Per capita annual health risks caused by metals in different types of groundwater by the drinking pathway/a⁻¹

二丰	井	水	泉	水	地丁	「河水
元素	成人	儿童	成人	儿童	成人	儿童
As	8. 00E - 08	8. 72E - 08	6. 05E – 07	6. 60E - 07	2. 89E - 07	3. 15E - 07
Cd	7.38E - 07	8.05E - 07	6.42E - 07	7.00E - 07	4. 50E - 07	4. 91E - 07
Cr	9.32E - 05	1.02E - 04	7.20E - 05	7.86E - 05	7.83E - 05	8. 55E - 05
Cu	1. 69E – 11	1.84E -11	5. 34E – 12	5. 83E – 12	2. 24E - 12	2. 44E - 12
Hg	1.85E - 10	2.01E - 10	1.08E - 10	1. 18E – 10	3. 41E - 09	3. 72E - 09
Pb	2. 49E – 10	2.72E - 10	2. 62E – 10	2. 86E – 10	1. 59E – 10	1. 74E – 10
Zn	3.07E - 10	3.35E - 10	7. 94E – 11	8. 66E – 11	6. 54E – 11	7. 13E – 11
Fe	9. 55E – 12	1.04E - 11	1.84E - 10	2.01E - 10	4. 59E – 12	5. 00E - 12
Al	1.78E - 11	1.94E – 11	1. 16E – 10	1. 26E – 10	5. 90E – 11	6. 43E – 11
Mn	2. 24E – 11	2.45E - 11	5. 60E – 10	6. 10E – 10	7. 94E – 11	8. 66E – 11
总风险	9. 40E - 05	1. 03E - 04	7. 33E - 05	7. 99E – 05	7. 91E - 05	8. 63E - 05

表 10 不同类型地下水金属元素经皮肤接触途径引起的个人平均年健康风险/a-1

Table 10	Per capita annual health risks caused by	metals in different types of groundwater by the	e dermal contact pathway/a -1

二丰	井	水	泉	水	地丁	河水
元素	成人	儿童	成人	儿童	成人	儿童
As	1. 82E - 09	1. 28E - 09	1. 38E - 08	9. 66E - 09	6. 57E - 09	4. 61E - 09
Cd	3.83E - 09	2.69E - 09	3.33E - 09	2.34E - 09	2. 33E - 09	1. 64E - 09
Cr	9.66E - 07	6.78E - 07	7.46E - 07	5.24E - 07	8. $12E - 07$	5.70E - 07
Cu	1.75E - 13	1. 23E – 13	5. 54E – 14	3.89E - 14	2. 32E - 14	1. 63E – 14
Hg	1.72E - 12	1.21E - 12	1.01E - 12	7.08E - 13	3. 18E – 11	2. 23E – 11
Pb	1. 72E – 14	1.21E - 14	1.81E - 14	1.27E - 14	1. 10E – 14	7. 72E – 15
Zn	2. 86E – 11	2.01E - 11	7. 40E – 12	5. 20E – 12	6. 10E – 12	4. 28E – 12
Fe	3.30E - 14	2. 31E – 14	6. 36E – 13	4. 46E – 13	1. 58E – 14	1. 11E – 14
Al	9. 22E – 13	6. 47E – 13	6. 00E – 12	4. 21E – 12	3.05E - 12	2. 14E – 12
Mn	2.97E - 13	2.09E - 13	7.41E - 12	5. 20E – 12	1. 05E – 12	7. 38E – 13
总风险	9.71E-07	6. 82E - 07	7. 64E - 07	5. 36E - 07	8. 21E - 07	5. 76E - 07

行更严格的控制. 从金属元素来说, 井水金属元素引起的健康风险大小顺序为: Cr > Cd > As > Zn > Pb > Hg > Mn > Al > Cu > Fe, 泉水为: Cr > Cd > As > Mn > Pb > Fe > Al > Hg > Zn > Cu, 地下河为: Cr > Cd > As > Hg > Pb > Mn > Zn > Al > Fe > Cu. 从致癌风险水平来说, 致癌总风险比非致癌总风险高4 ~6 个数量级, 表明研究区地下水的健康总风险主要来源于致癌性金属元素, 特别是 Cr, 致癌风险的分布代表了研究区的健康风险格局, 此结果与王若师等[37] 对东江流域典型村饮用水源地重金属污染健康风险评价一致, 因此应将 Cr 作为风险决策管理重点.

3 结论

- (1) 研究区井水的 pH 值、TDS、总硬度 (CaCO₃)、NO₃⁻ 和 Na⁺平均质量浓度最高,泉水的高锰酸盐指数、SO₄²、Cl⁻、F⁻和 NH₄⁺ 最高,地下河水的 NO₂⁻ 最高. 按金属元素平均质量浓度堆积总浓度大小顺序为泉水 > 井水 > 地下河水. 井水中 NO₃⁻、Zn 和 Hg 超过 III 类水质标准限值,泉水中 NO₃⁻、F⁻、NH₄⁺、高锰酸盐指数、Fe、Mn 和 Al 超标,地下河水 NO₃⁻、NO₂⁻ 和 Hg 超标. 井水、泉水和地下河水的综合评价分值分别为 7. 10、7. 10 和 4. 26,水质级别均为较差级别,按水质级别为良好的样品数百分比大小顺序为地下河水 (36. 36%) > 井水 (35. 71%) > 泉水 (33. 33%).
- (2)研究区地下水的 pH 值影响着 Cu 和 As 质量浓度的分布,高硬度和高矿化度利于 Cr 富集,不利于 Fe、Mn 和 Al 富集,还原环境(高锰酸盐指数、 NH_4^+ 和 F^-)利于 As 富集. 地下水 Zn、Pb、Cd 和 Cu 经过的环境地球化学作用相似,Fe、Al 和 Mn 来源相似,As 和 Hg 可能具有相似的空间变化规律,Cr 与其它金属元素来源不同或环境地球化学性质

不同.

- (3)研究区3种类型地下水金属元素引起的健康总风险均高于最大可接受风险水平(5.0×10⁻⁵ a⁻¹),总风险大小顺序为井水>地下河水>泉水. 致癌总风险(高于5.0×10⁻⁵ a⁻¹)比非致癌总风险(小于10⁻⁶ a⁻¹)高4~6个数量级,健康总风险主要来源于致癌性金属元素 Cr, Cr 致癌风险值高于5.0×10⁻⁵ a⁻¹,应将 Cr 作为风险决策管理重点. 儿童总风险均大于成人,经饮水途径引起的健康风险比皮肤接触途径高2~3个数量级,饮水途径是金属元素的主要暴露途径,应在饮水安全方面对儿童进行更严格控制. 井水、泉水和地下河水的致癌总风险来源物质顺序均为 Cr > Cd > As,非致癌总风险来源物质质大小顺序分别为 Zn > Pb > Hg > Mn > Al > Cu > Fe、Mn > Pb > Fe > Al > Hg > Zn > Cu 和 Hg > Pb > Mn > Zn > Al > Fe > Cu.
- (4)结合内梅罗综合指数法和健康风险评价, 地下河水的综合评价分值最低,健康风险高于泉水,泉水的健康风险最低,水质指标超过率最高, Cr元素致癌风险最高.从饮水安全考虑,需特别注 意在饮用前对井水、地下河水和泉水进行 Cr污染物控制,适当处理后可作为生活饮用水.

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Migration Characteristics of Manganese During Rainfall Events and Its Impacts on Water Quality in a Drinking Water Source Reservoir	(2730) (2738) (2745) (2745) (2753) (2764) (2773) (2783) (2800) (2807) (2813) (2821) (2827) (2840) (2847) (2858) (2859)
Migration Characteristics of Manganese During Rainfall Events and Its Impacts on Water Quality in a Drinking Water Source Reservoir	(2730) (2738) (2745) (2745) (2753) (2764) (2773) (2783) (2793) (2800) (2807) (2813) (2821) (2827) (2840) (2847) (2858) (2859)
Migration Characteristics of Manganese During Rainfall Events and Its Impacts on Water Quality in a Drinking Water Source Reservoir	(2730) (2738) (2745) (2745) (2753) (2764) (2773) (2783) (2793) (2800) (2807) (2813) (2821) (2827) (2840) (2847) (2858) (2859)
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Migration Characteristics of Manganese During Rainfall Events and Its Impacts on Water Quality in a Drinking Water Source Reservoir	(2730) (2738) (2745) (2745) (2753) (2764) (2773) (2783) (2800) (2807) (2813) (2821) (2827) (2840) (2847) (2858) (2869) (2877) (2858) (2879) (2879) (2871) (2871) (2871) (2872) (2873) (2873) (2873) (2873) (2874) (2874) (2874) (2875) (2
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