Spatiotemporal Distributions of Fluorine and Arsenic in Rivers With the Role of Mining Industry and Related Human Health Risk Assessments in Kyrgyzstan

Yizhen Li
Xinjiang Institute of Ecology and Geography
Long Ma (malong@ms.xjb.ac.cn)
Xinjiang Institute of Ecology and Geography
Jilili Abuduwaili
Xinjiang Institute of Ecology and Geography
Yaoming Li
Xinjiang Institute of Ecology and Geography
Salamat Abdyzhapar uulu
Institute of Geology, National Academy of Sciences of Kyrgyzstan

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Spatiotemporal distributions of fluorine and arsenic in rivers with the role of mining industry and related human health risk assessments in Kyrgyzstan

Yizhen Li[1,2,3], Long Ma[1,2,3]*, Jilili Abuduwaili[1,2,3], Yaoming Li[1,2,3], Salamat Abdyzhapar uulu[2,4]

[1] State Key Laboratory of Desert and Oasis Ecology, Xinjiang Institute of Ecology and Geography, Chinese Academy of Sciences, Urumqi 830011, China

[2] Research Center for Ecology and Environment of Central Asia, Chinese Academy of Sciences, Urumqi 830011, China

[3] University of Chinese Academy of Sciences, Beijing 100049, China

[4] Institute of Geology, National Academy of Sciences of Kyrgyzstan, Bishkek 720461, Kyrgyzstan;

*** Correspondence to malong@ms.xjb.ac.cn.**

Abstract

To determine the spatiotemporal distributions and human health risks of fluoride and arsenic in the rivers of Kyrgyzstan as influenced by mining and other industries, 169 water samples were collected from the main rivers and tributaries of Kyrgyzstan from 2016 to 2018. Through the use of cold and hot spot analysis, multivariate statistical analysis and health risk assessment model, the results indicated that the fluorine and arsenic concentrations in river waters increased year by year from 2016 to 2018. In total, 2.38%, 3.26% and 10.64% of the analyzed samples exceeded the drinking water standard of a maximum permissible limit of 1 mg/L for fluoride, and 0%, 1.09% and 2.13% of the samples exceeded the limit of 10 μg/L for arsenic in 2016, 2017 and 2018, respectively. The gathering areas for high fluorine concentrations were mainly distributed in the Issyk-Kul Basin, Chu River Valley and Fergana

Basin, and the gathering areas for high arsenic concentrations were mainly concentrated in the Chu River Valley and southern Fergana Basin. Although fluorine and arsenic were not found to exceed the limits simultaneously, the two pollutants accumulated high values in the southern Fergana Basin in 2018, which indicated the risk of joint poisoning. The distributions of high fluorine and arsenic were found to be determined by mining, industrial and agricultural activities, but not by natural sources. From 2016 to 2018, arsenic concentrations in the river water of Kyrgyzstan created a high risk of carcinogenesis by the ingestion intake exposure route, which resulted in the total risk of health hazards to children and adults caused by fluoride and arsenic to exceed the maximum acceptable ranges. Therefore, further monitoring and management are urgently needed.

Keywords

Fluoride; arsenic; spatiotemporal distribution; health risk; Kyrgyzstan

Introduction

Fluorine and arsenic are both trace elements that exist widely in nature. Fluoride can promote bone and tooth development, but excessive exposure to high fluoride concentrations of fluoride in drinking water can lead to dental fluorosis and skeletal fluorosis [1, 2]. Arsenic is considered to be an element that is harmful to human health, and arsenic intake can increase the risk of bladder cancer, liver cancer, kidney cancer and skin cancer and can lead to congestive heart failure[3]. In addition, studies have shown that arsenic and fluorine have combined toxicity to the human body [4], and the combined effect of arsenic and fluorine also affects children's intelligence, growth and development [5]. Due to the dual interference of natural factors and human activities, fluoride and arsenic in the water bodies of many countries in the world have exceeded the established limits, which have aroused widespread concern[6-8].

Kyrgyzstan is located in the main part of the "Central Asian orogenic belt". The Tianshan metallogenic belt in Kyrgyzstan is the core of the Central Asian metal mineral resource base, which is rich in mineral resources. With the introduction of international investment and related policies [9-11], the mining industry, as one of the emerging industries in Kyrgyzstan, began to develop rapidly. However, the Tianshan Mountains in Kyrgyzstan are not only the base of mineral resources but are also the "water tower" of all of Central Asia [12], where most Central Asian rivers originate. Therefore, the mining industry and its associated processing industries have become potential threats to water quality guarantees and water resource security in Central Asia. At present, some scholars have analyzed the arsenic or fluorine concentrations in some rivers of Kyrgyzstan, such as the Shu River, Shor-Koo River, and Aksu River [13], surface water in Mailuu Suu town [14, 15] and the Naryn River [16]. Simultaneously, some scholars have conducted human health risk assessments for potential toxic elements in the rivers of Kyrgyzstan and have found that the arsenic levels in the rivers of the Issyk−Kul Basin [17] and the transboundary river of the Chu-Talas River in Kyrgyzstan [18] were harmful to human health. However, the above research objects were generally limited to a single river, watershed or prefecture, and the concentrations and distributions of fluorine and arsenic in the rivers of Kyrgyzstan, spatiotemporal distribution characteristics of fluorine and arsenic in river water, influencing factors of spatial distributions of fluorine and arsenic in river waters, whether fluorine and arsenic simultaneously exceeded the established limits, and the health risk of both to human health are still unclear. To provide a theoretical basis for national water resource management, water pollution control, public health protection and policy makers in the relevant departments, it is urgent to determine the temporal and spatial distributions of fluorine and arsenic in the river waters of Kyrgyzstan and conduct related human health risk assessments.

Based on this, a total of 169 samples were collected from the main rivers and their tributaries in Kyrgyzstan from 2016 to 2018. The concentrations and spatiotemporal distribution characteristics of fluorine and arsenic in river waters were analyzed, and human health risks were evaluated for the ingestion and dermal exposure routes of fluorine and arsenic for both adults and children in this study. The results can provide a reference for water resource management and sustainable utilization in Kyrgyzstan and Central Asia.

Geographical Setting

Kyrgyzstan is located in the hinterland of Eurasia and is one of five Central Asian countries (Figure 1a); its population is slightly greater than 6 million [19]. As a typical mountainous country, 94% of Kyrgyzstan's territory is located higher than 1,000 meters above sea level [20], and lowlands only account for 15% of its land area, which are mainly distributed in the Fergana basin in the southwest and the Taras and Chu River Valleys in the north (Figure 1b). Kyrgyzstan is divided into seven states: Chuy, Talas, Osh, Jalal-Abad, Naryn, Ysykt and Batken (Figure 1c). Due to the presence of large glaciers in its alpine regions, Kyrgyzstan has some of the richest surface water resources in Central Asia. There are 73 rivers with lengths greater than 50 km, including the Naryn River, Chu River, Syr Darya, Karadarya River, Tarim River and Sarezaz River [21]. Kyrgyzstan has rich gold, copper, antimony, mercury, uranium, and other heavy rare earth material deposits [22]. In recent years, under the background of "one belt, one road", Kyrgyzstan's industry has grown rapidly, and its commodity trade volume has also increased year by year, among which gold is its primary export commodity [23]. Kyrgyzstan has also become a country with the most obvious effects from promoting domestic economic development with the development of gold production.

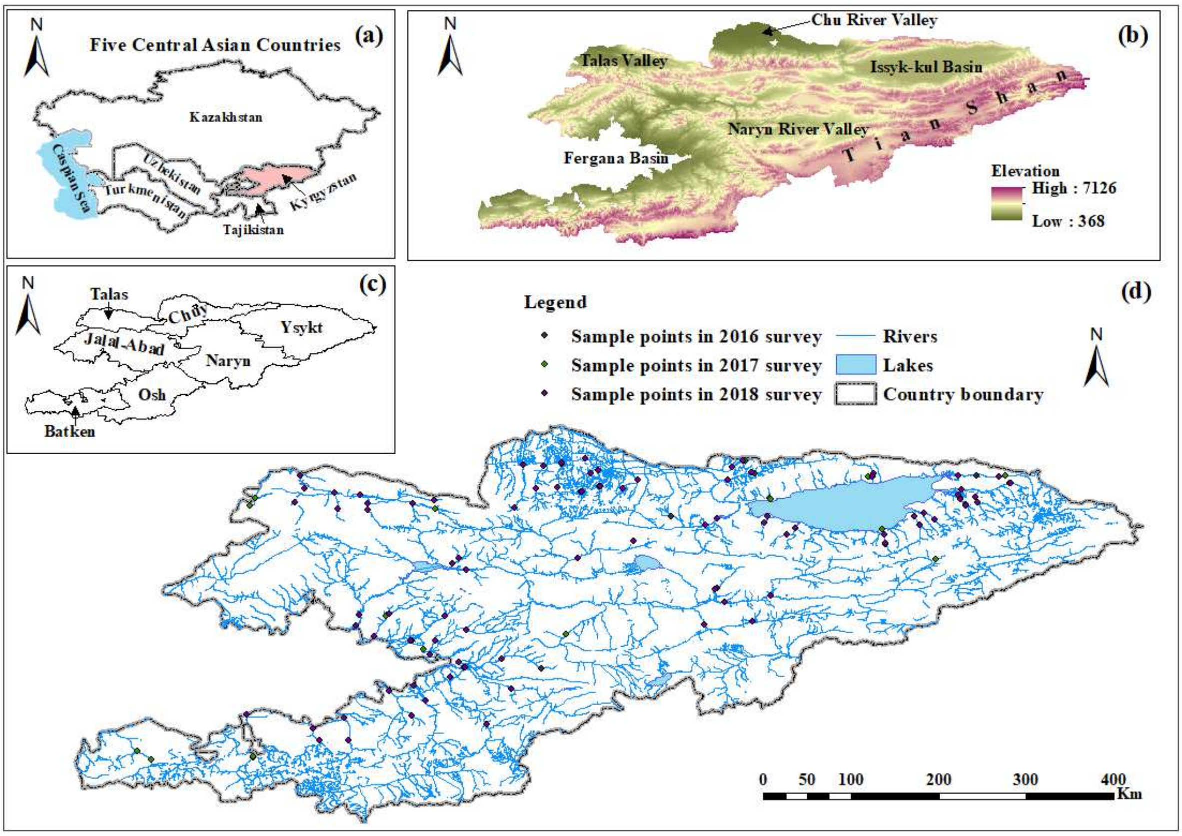

Figure 1 Location of the study area

Results

Temporal variations in fluorine and arsenic in the rivers of Kyrgyzstan

The results from determining fluorine and arsenic concentrations in the river samples of Kyrgyzstan from 2016 to 2018 are shown in Table 1. The fluorine concentrations in surface water ranged from 0.11 to 1.23 mg/L in 2016, from 0.22 to 2.26 mg/L in 2017 and from 0.09 to 1.6 mg/L in 2018. In general, the fluorine contents of the rivers in Kyrgyzstan mostly exceeded the average fluoride concentration (0.1 mg/L) of rivers worldwide [24]. According to the statistics (Figure S1 and Table 1), 2.38%, 10.71% and 3.26% of the samples in the river waters of Kyrgyzstan exceeded the Chinese drinking water standard (1 mg/L), and 5.43%, 10.64% and 13.83% of the samples exceeded the Japanese drinking water standard (0.8 mg/L) from 2016 to 2018, respectively. These results indicated that the surface waters in some areas of Kyrgyzstan cannot be directly used as drinking water in terms of the fluoride concentrations. The ranges of arsenic concentrations in the surface waters of Kyrgyzstan from 2016 to 2018 ranged from 0.08 to 8.3 μg/L, 0.18 to 16.53 μg/L and 0.15 to 22.04 μg/L, respectively. The maximum arsenic concentrations increased year by year from 2016 to 2018. According to the World Health Organization (WHO) guidelines for drinking water quality, arsenic concentrations in all samples of surface water in Kyrgyzstan did not exceed the limit of 10 μg/L in 2016, and the arsenic concentrations in one (1.09%) and two (2.13%) samples of surface water in 2017 and 2018 exceeded the limit, respectively. Arsenic concentrations not only showed a trend of increasing year by year in their peak values but also increased year by year in the number sampling points that exceeded the limit.

Table 1 Descriptive statistical characteristics of fluorine and arsenic in the rivers of Kyrgyzstan from 2016 to 2018

Parameter		Fluorine concentration (mg/L)			Arsenic concentration (ug/L)		
		2016	2017	2018	2016	2017	2018
Range		1.13	2.04	1.51	8.22	16.35	21.89
Minimum		0.11	0.22	0.09	0.08	0.18	0.15
Maximum		1.23	2.26	1.60	8.30	16.53	22.04
Mean		0.47	0.47	0.52	1.20	1.56	1.74
Standard deviation		0.26	0.26	0.33	1.34	2.18	2.79
Overlimit rate	(1 mg/L)	2.38%	3.26%	10.64%	0.00%	1.09%	2.13%
	(0.8 mg/L)	10.71%	5.43%	13.83%			

Spatial variations in fluorine and arsenic in the rivers of Kyrgyzstan

To analyze the specific locations with excessive fluoride and arsenic concentrations in surface water, Figure 2 shows the spatial distributions of fluoride and arsenic in the surface waters of

Kyrgyzstan from 2016 to 2018. In 2016, high concentrations of fluorine were only distributed in the Issyk−Kul Basin. After 2017, fluorine concentrations in the Chu River Valley and Fergana Basin (Figure 1b) exceeded the limit. By 2018, the number of samples in which fluorine concentrations exceeded the limit in the Issyk−Kul Basin (Figure 1b) increased, and the number samples in which the fluorine concentrations exceeded the limit also increased in the southern Fergana Basin (Batken state, Figure 1). The results showed that most fluoride in the surface waters of Kyrgyzstan was concentrated in low valleys and basins, and the locations and degrees of excessive fluoride concentrations increased. In 2017 and 2018, arsenic in surface water exceeded the limit in the Ak-Suu reach of the Chu River Valley, and the degree of exceedance in 2018 was more serious than in 2017.

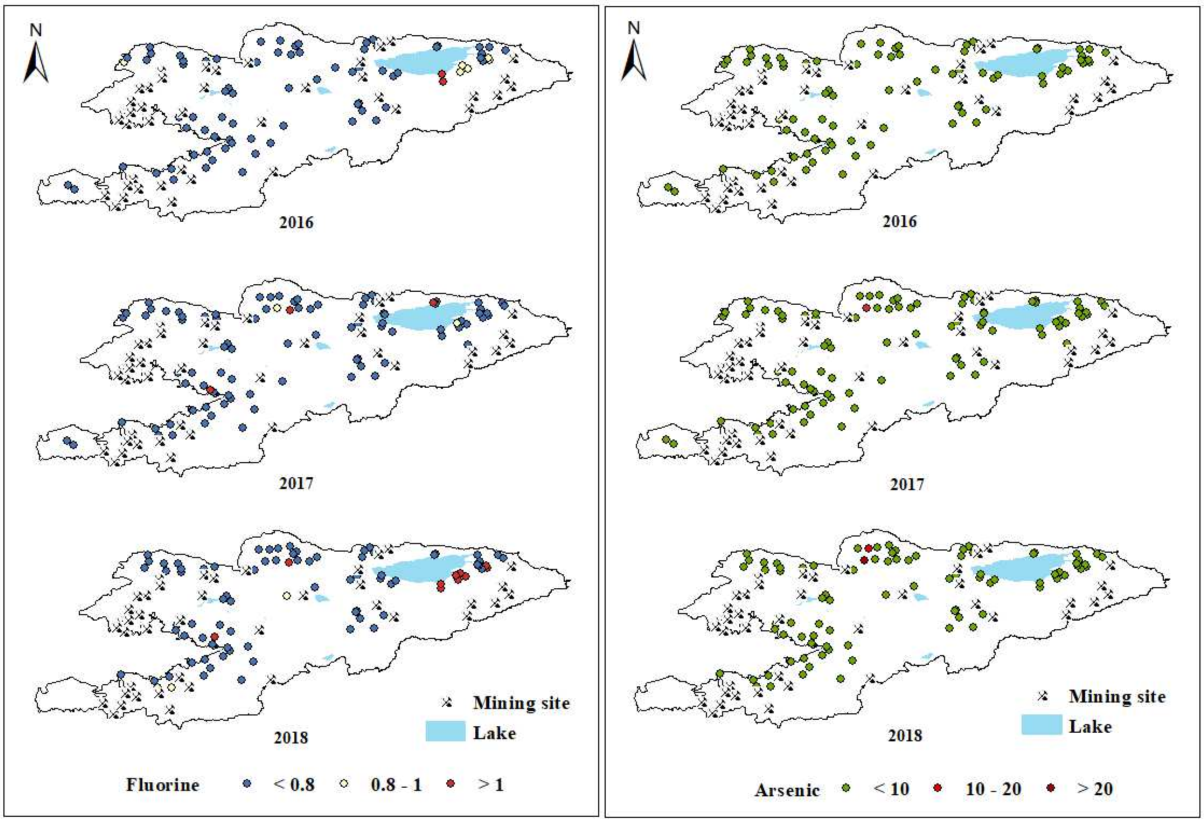

Figure 2 Spatial distributions of fluorine and arsenic in the surface waters of Kyrgyzstan from 2016 to 2018

The above analysis only focused on the spatiotemporal distribution characteristics of excessive fluoride and arsenic. However, understanding the overall distribution and high- and low-value aggregation distribution characteristics of fluoride and arsenic is also very important for preventing and controlling pollution. Therefore, the distributions of fluorine and arsenic in the surface waters of Kyrgyzstan from 2016 to 2018 were analyzed by using the Getis-Ord Gi^* index in ArcGIS, and the sampling points of fluorine and arsenic were divided into three types in this study: cold points, hot points and random points. The results are shown in Figure 3.

According to the statistics obtained from ArcGIS (Table S4), fluorine cold points in surface water accounted for 19.75%, 9.78% and 22.58% of the sampling points, random points accounted for 64.20%, 82.61% and 54.84%, and hot points accounted for 16.05%, 7.61% and 22.58% from 2016 to 2018, respectively. It can be seen that the cold and hot spots for fluorine exhibited a pattern of "decrease-increase" in these three years, and the randomness of fluorine distributions in the surface waters of Kyrgyzstan was highest in 2017, while the gathering points of significant high values (hot spots) and significant low values (cold spots) of fluorine in 2018 were more numerous than those in 2016, which indicated that the randomness of fluorine distributions gradually decreased and that the aggregation distribution increased. From a spatial perspective, the distribution of fluorine cold spots in surface water showed a pattern of moving from the east of the Fergana Basin to the Talas Valley, and the hot spots are mainly located at the southern edge of the Issyk−Kul Basin. By 2018, fluorine hot spots also appeared in the southern Fergana Basin. From 2016 to 2018, only in 2016 (3.61%) were the arsenic cold spots distributed in the eastern Fergana basin, while in 2017 and 2018, there were no arsenic cold spots. The totals of the hot spots accounted for 18.07%, 16.30% and 15.05% from 2016 to 2018, respectively, and exhibited a decreasing trend year by year, but the proportion of hot spots that passed the 0.01 significance level increased year by year. In 2016, arsenic hot spots were mainly distributed in the middle and eastern Chu River Valley and southern Fergana Basin. By 2018, the number and significance of arsenic hot spots gradually became concentrated in the middle of the Chu River Valley.

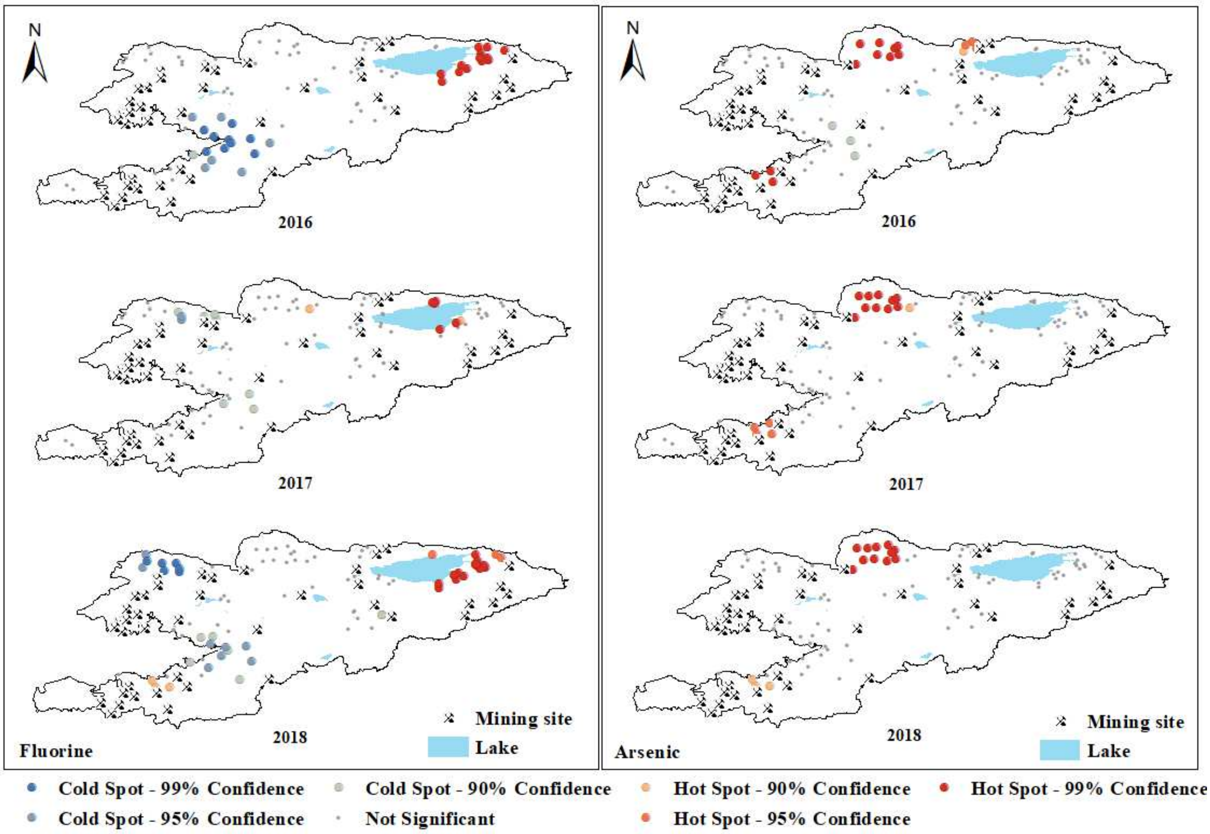

Figure 3 Distributions of cold and hot spots of fluorine and arsenic in the surface waters of Kyrgyzstan from 2016 to 2018

When examining Figures 2 and 3, it is evident that fluorine and arsenic at all sampling points did not simultaneously exceed the standards from 2016 to 2018. However, from the accumulation of high-concentration points (hot spots), both pollutants accumulated high values in the southern Fergana Basin (Batken state) in 2018, which demonstrated a potential risk that fluorine and arsenic exceeded the standard at the same time. Therefore, it is suggested that fluoride and arsenic monitoring be strengthened in the rivers in this area and that corresponding measures be introduced to reduce the risk of combined fluoride and arsenic poisoning.

Health risk assessment of arsenic and fluorine in the river waters of Kyrgyzstan

The annual noncarcinogenic risk, carcinogenic risk and total risk assessment results for fluoride and arsenic in Kyrgyz rivers that were caused by ingestion intake and dermal exposure routes from 2016 to 2018 are shown in Table 2, Table 3 and Figure 4. Table 2 shows that the noncarcinogenic risk of fluoride and arsenic in the surface waters of Kyrgyzstan for adults and children from 2016 to 2018 was lower than the maximum acceptable risk level of $5\times10^{-5}a^{-1}$, which has been recommended by the International Commission on Radiation Protection (ICRP). For different risk-bearing individuals, the noncarcinogenic risk of fluoride and arsenic for children was higher than that for adults on the whole, which indicates that children are more sensitive risk receptors than adults and had higher exposures to fluoride and arsenic; this result is consistent with previous research results [2, 8]. For different pollutants, the noncarcinogenic risk of fluoride through ingestion intake was less than that through dermal exposure, while the noncarcinogenic risk of arsenic through ingestion intake was greater than that through dermal exposure, which indicated that dermal contact was the main exposure route for noncarcinogenic fluoride risk, and ingestion intake was the main exposure route for noncarcinogenic arsenic risk. For different years, the noncarcinogenic risks (mean values) of fluoride and arsenic for adults and children gradually increased from 2016 to 2018, which indicated that the noncarcinogenic risk of fluoride and arsenic to the human body increased during this period.

Table 3 shows that the average carcinogenic risks of adults and children caused by arsenic through ingestion intake were higher than the maximum acceptable risk level of carcinogens, as recommended by the Swedish Environmental Protection Agency and Dutch Ministry of Construction and Environment of $1.0\times10^{-6}a^{-1}$. According to the statistics (Figure 4), more than 89% of all samples exceeded the standard. However, the carcinogenic risk that was caused by the dermal exposure route was less than the maximum acceptable risk level and was 5-6 orders of magnitude lower than that caused by the ingestion intake route, which indicated that the ingestion intake route was the main exposure route for arsenic carcinogenic risk. From the perspective of different receptors, the carcinogenic risk for children was higher than that for adults, and the percentage of samples with excessive carcinogenic risk that was caused by ingestion intake was also higher than that of adults, which indicated that children were also more sensitive risk receptors in terms of their carcinogenic risk. In terms of time, the average carcinogenic risk that was caused by arsenic increased gradually from 2016 to 2018, but the percentage of samples with excessive carcinogenic risk caused by ingestion intake decreased year by year, which indicated that the carcinogenic risk from arsenic in the surface waters of Kyrgyzstan has been improved to a certain extent, but control of carcinogenic risk levels needs to be further strengthened.

Table 2 Noncarcinogenic risks of fluoride and arsenic in surface waters through ingestion and dermal exposure routes / a^{-1}

Year	Receptors	Ingestion intake route (fluorine)			Dermal exposure route (fluorine)			Ingestion intake route (arsenic)			Dermal exposure route (arsenic)		
		Minimum	Maximum	Mean	Minimum	Maximum	Mean	Minimum	Maximum	Mean	Minimum	Maximum	Mean
2016	Adults	6.45×10^{-10}	7.34×10^{-9}	2.79×10^{-9}	1.26×10^{-9}	1.44×10^{-8}	5.46×10^{-9}	9.37×10^{-11}	9.88×10^{-9}	1.43×10^{-9}	9.17×10^{-13}	9.67×10^{-11}	1.4×10^{-11}
	Children	1.2×10^{-9}	1.37×10^{-8}	5.2×10^{-9}	2.1×10^{-9}	2.39×10^{-8}	9.09×10^{-9}	1.75×10^{-10}	1.84×10^{-8}	2.66×10^{-9}	1.53×10^{-12}	1.61×10^{-10}	2.32×10^{-11}
2017	Adults	1.31×10^{-9}	1.35×10^{-8}	2.81×10^{-9}	2.57×10^{-9}	2.64×10^{-8}	5.51×10^{-9}	2.13×10^{-10}	1.97×10^{-8}	1.86×10^{-9}	2.08×10^{-12}	1.93×10^{-10}	1.82×10^{-11}
	Children	2.45×10^{-9}	2.51×10^{-8}	5.25×10^{-9}	4.29×10^{-9}	4.39×10^{-8}	9.17×10^{-9}	3.97×10^{-10}	3.67×10^{-8}	3.46×10^{-9}	3.47×10^{-12}	3.21×10^{-10}	3.02×10^{-11}
2018	Adults	5.2×10^{-10}	9.5×10^{-9}	3.12×10^{-9}	1.02×10^{-9}	1.86×10^{-8}	6.1×10^{-9}	1.8×10^{-10}	2.62×10^{-8}	2.07×10^{-9}	1.76×10^{-12}	2.57×10^{-10}	2.03×10^{-11}
	Children	9.7×10^{-10}	1.77×10^{-8}	5.82×10^{-9}	1.69×10^{-9}	3.1×10^{-8}	1.02×10^{-8}	3.36×10^{-10}	4.9×10^{-8}	3.87×10^{-9}	2.93×10^{-12}	4.28×10^{-10}	3.38×10^{-11}

Table 3 Carcinogenic and total risks of fluoride and arsenic in surface waters by ingestion and dermal exposure routes / a^{-1}

Year	Receptors	Ingestion intake route (arsenic)			Dermal exposure route (arsenic)			Total risk
		Minimum	Maximum	Mean	Minimum	Maximum	Mean	
2016	Adults	4.22×10^{-7}	4.44×10^{-5}	6.41×10^{-6}	9.17×10^{-13}	9.67×10^{-11}	1.4×10^{-11}	6.422×10^{-6}
	Children	7.87×10^{-7}	8.28×10^{-5}	1.2×10^{-5}	1.53×10^{-12}	1.61×10^{-10}	2.32×10^{-11}	1.198×10^{-5}
2017	Adults	9.57×10^{-7}	8.83×10^{-5}	8.34×10^{-6}	2.08×10^{-12}	1.93×10^{-10}	1.82×10^{-11}	8.35×10^{-6}
	Children	1.79×10^{-6}	1.64×10^{-4}	1.56×10^{-5}	3.47×10^{-12}	3.21×10^{-10}	3.02×10^{-11}	1.558×10^{-5}
2018	Adults	8.09×10^{-7}	1.18×10^{-4}	9.33×10^{-6}	1.76×10^{-12}	2.57×10^{-10}	2.03×10^{-11}	9.339×10^{-6}
	Children	1.51×10^{-6}	2.19×10^{-4}	1.74×10^{-5}	2.93×10^{-12}	4.28×10^{-10}	3.38×10^{-11}	1.741×10^{-5}

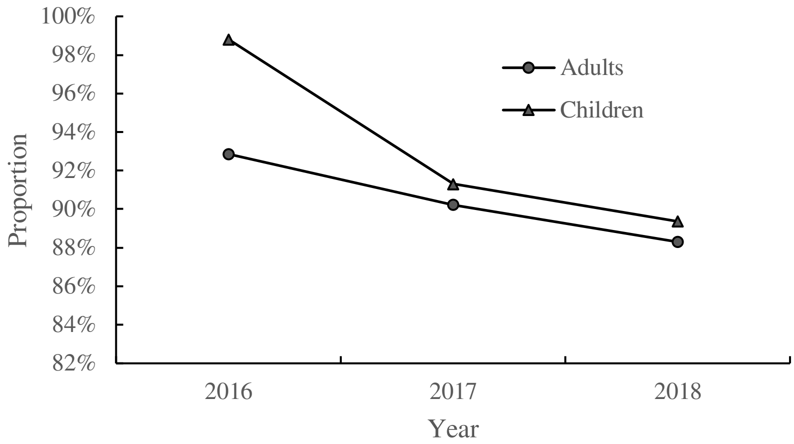

Figure 4 Proportions of carcinogenic risk caused by arsenic through ingestion intake which exceeds the maximum acceptable risk level

From 2016 to 2018, the total risks of health hazards to children and adults that were caused by fluoride and arsenic in the surface waters of Kyrgyzstan were 1.198×10^{-5}~$1.741\times10^{-5}a^{-1}$ and 6.422×10^{-6}~$9.339\times10^{-6}a^{-1}$, respectively, which were both higher than the maximum acceptable risk level of $1.0\times10^{-6}a^{-1}$, which indicated that the water bodies posed health risks.

Discussion

Influencing factors of fluorine and arsenic distributions in the study area

Fluorine in river water may come from both natural sources and anthropogenic activities. Natural sources include the erosion and leaching of fluorine-rich soil and rock[2, 25], deposition of fluorine-containing dust in the atmosphere and transport by precipitation [24]. Anthropogenic activities include metallurgy, ceramics, coal-fired power generation, metal mining and smelting, electrical and electronic industries, textile dyeing, polytetrafluoroethylene plastics, phosphate mining and phosphorus-containing fertilizers, fluorine-containing pesticides, toothpaste and fire extinguishing agents, and air conditioning refrigerants (CFCs) [2, 26-28]. Similarly, the arsenic sources in rivers can be divided into natural and anthropogenic activities. The natural factors mainly include rock weathering, soil erosion and volcanic eruptions, while the anthropogenic factors mainly include mining of arsenic-bearing deposits, gold mining, arsenic-containing pesticides, and discharge of industrial arsenic-containing wastewater [29-31]. In the study area, the Kyrgyz industries mainly include mining, electric power, fuel, chemical industries, food industries, metals, machinery manufacturing, building materials, glass and ceramics, which are mainly distributed in Chuy and Osh states; agriculture is mainly distributed in Chuy, Osh, Jalalabad and Ysykt states [32].

To understand the homology of fluoride and arsenic with the main ions in Kyrgyz River waters, Pearson correlation analysis and principal component analysis were conducted between fluoride and arsenic with the main ions in the Kyrgyz River water from 2016 to 2018 by using SPSS 25 statistical analysis software. The results are shown in Tables 4 and 5. Table 4 shows that the correlation coefficients (absolute values) between fluorine and arsenic and the water chemical parameters were not above than 0.4 and that the correlations between fluorine and arsenic and various ionic components were different in different years. Therefore, it can be inferred that the fluorine and arsenic in the river waters of Kyrgyzstan from 2016 to 2018 were mainly affected by human activities and that natural sources that were controlled by the geological background made little contributions to the distribution characteristics of fluorine and arsenic.

Table 4 Correlations between fluorine, arsenic and the physicochemical indexes of the water environment

Year	2016		2017		2018	
	Arsenic	Fluorine	Arsenic	Fluorine	Arsenic	Fluorine
HCO_3^-	**-0.285****	-0.189	0.028	-0.060	-0.040	-0.164
Cl^-	-0.156	-0.032	0.036	**0.240***	0.014	0.181
SO_4^{2-}	0.077	-0.066	0.112	**0.231***	0.120	**0.205***
NO_3^-	-0.092	-0.165	0.089	0.154	0.026	-0.076
K^+	-0.032	0.104	0.029	0.194	0.025	**0.302****

Na^+	-0.137	-0.115	0.042	**0.252***	0.062	0.201
Ca^{2+}	-0.032	0.081	**0.365****	0.121	0.160	-0.010
Mg^{2+}	**-0.230***	**-0.225***	0.056	0.127	0.063	0.129

Through data tests, the KMO values of the three-year data were 0.639, 0.735 and 0.647, and Bartlett values were all 0, which indicated that the samples met the requirements of a reasonable data distribution and could be further analyzed by principal component analysis. The results showed that the main ions in the river samples of Kyrgyzstan from 2016 to 2018 were in the 1st principal component (except HCO_3^- in 2017), and fluorine and arsenic were in the 2nd and 3rd principal components, respectively, which indicated that the water chemical parameters in the river samples belonged to one category, while fluorine and arsenic belonged to two other categories. The results of principal component analysis showed that the sources of fluorine and arsenic were different from those of the main ions in the water environment, which also verified the conclusion that was inferred from the correlation analysis results and further indicated that the anthropogenic sources of fluorine and arsenic in river water were also different.

Table 5 Principal component loadings of rivers in Kyrgyzstan from 2016 to 2018

Indicators	2016			2017			2018		
	PCA1	PCA2	PCA3	PCA1	PCA2	PCA3	PCA1	PCA2	PCA3
HCO_3^-	**0.76**	-0.32	-0.16	0.47	**0.71**	-0.28	**0.70**	-0.50	-0.17
F	-0.12	**0.68**	**-0.57**	0.23	-0.45	0.48	0.16	**0.83**	-0.05
Cl^-	**0.78**	-0.04	-0.31	**0.93**	-0.30	-0.01	**0.95**	0.11	-0.05
SO_4^{2-}	**0.75**	0.35	0.29	**0.97**	-0.17	0.03	**0.95**	0.17	0.10
NO_3^-	0.47	-0.18	0.46	**0.77**	0.14	-0.03	0.53	-0.44	-0.13
K^+	**0.77**	0.27	-0.09	**0.96**	-0.15	-0.07	**0.91**	0.19	-0.06
Na^+	**0.89**	-0.05	-0.11	**0.95**	-0.28	-0.01	**0.95**	0.18	0.02
Ca^{2+}	**0.83**	0.31	0.04	**0.83**	0.46	0.13	**0.81**	-0.27	0.12
Mg^{2+}	**0.88**	-0.13	0.10	**0.95**	0.08	-0.13	**0.96**	0.06	0.02
As	-0.19	**0.66**	0.49	0.13	0.43	**0.84**	0.08	-0.08	**0.98**
Characteristic value	4.87	1.35	1.01	6.10	1.34	1.06	5.91	1.33	1.04
Variance /%	48.70	13.52	10.13	61.05	13.40	10.55	59.15	13.34	10.38
Cumulative variance /%	48.70	62.22	72.35	61.05	74.45	85.00	59.15	72.48	82.86

When combined with the distributions of mineral resources[22] and distribution characteristics of fluorine and arsenic in Kyrgyzstan (Figure 2 and Figure 3), it is apparent that most of the high fluorine concentrations were concentrated in the Issyk-Kul Lake Basin, and a small amount of fluorine was concentrated in the Fergana Basin (in Batken state). There are abundant mineral resources in the mountain areas of the upper reaches of the river in the Issyk-Kul Basin and Batken state, where the Kumtor gold mine (the largest gold mine in Kyrgyzstan) and many other metal mining sites are located. Intensive mining may cause fluorine accumulations in the river, and high fluorine concentrations may be attributed to metal mining and smelting. In addition, Ysykt state is also the main agricultural area in Kyrgyzstan, and fluorine-containing pesticides are also one of the sources of high fluorine concentrations in this area. The sampling sites that had arsenic concentrations which exceeded the standard were mainly distributed in the dense, urban area of the Chu River Valley, which may be related to discharges of industrial arsenic-containing wastewater in the city. In addition, agricultural activities in Chu state that caused arsenic pesticide pollution were also one of the contributors to the excessive arsenic levels in this area. Simultaneously, analysis of cold and hot spots showed that, in addition to the Chu River Valley, high-value gathering points (hot spots) were also present in the southern Fergana Basin, which indicated that the mining industry in this area may pose potential threats due to excessive arsenic levels.

Uncertainty analysis of health risk assessment and management of fluorine and arsenic

Human environmental health risk assessments are an important auxiliary method in any decision-making process, which aim to minimize the impact of human activities on the environment [33]. In this study, the risks of fluoride and arsenic in Kyrgyz River water to human health were quantitatively evaluated, but there are still many uncertainties in the analysis process. First, in the selection of the mathematical model and its parameters [34], the mathematical model is a function of multiple variables [33], and the parameters used in this study were taken directly from the relevant literature, which increased the uncertainty of the evaluation results to a certain extent. Moreover, simultaneous exposure to fluoride and arsenic may cause joint toxicity, and this joint toxicity may have synergistic or antagonistic effects on different organs and systems of the human body [35, 36], which increases the errors in health risk assessment results for a single element. Finally, only the health risks caused by drinking water and skin exposure to fluoride and arsenic were considered in this study; however, pollutants can also be harmful to human health through other exposure routes, such as oral and nasal inhalation [37] and direct contact [38]. In addition, the surface water resources in Central Asia are used not only for direct drinking water and entertainment water but also for farmland irrigation [17, 39]. Fluorine and arsenic in water can be absorbed through plant roots and leaves and then enter the human body via the food chain, which increases the risk of harm to human health. Therefore, in view of the above uncertain factors, error control of health risk assessments that relate to fluoride and arsenic needs to be further studied.

Water pollution is the most prominent problem for water resource security. To achieve sustainable management and utilization of water resources, the water quality security of river water bodies should be highly valued. According to the results of the health risk assessment, the total risk of health hazards to children and adults that is caused by fluoride and arsenic in the surface waters of Kyrgyzstan exceeded the maximum acceptable range from 2016 to 2018, so it is urgent to control fluoride and arsenic concentrations in river water and formulate relevant management policies. In terms of pollution sources, the mining industry and its associated processing industries in Kyrgyzstan present potential threats of fluoride and arsenic pollution in river water. It is necessary to strengthen the monitoring of pollutants that are exported by the mining industry and other industries to the environment and to specify appropriate restrictions and regulations. Chemical fertilizers and pesticides from agricultural activities are also sources of fluorine and arsenic pollution, so measures should be taken to avoid the excessive use of chemical fertilizers and pesticides. In terms of water supply, water quality improvement projects are the fundamental measure to prevent and control arsenic poisoning and fluorosis in drinking water [40]. At present, there are mature theories and methods that can simultaneously remove fluoride and arsenic from water, such as the membrane method [41], adsorption method [42] and ion exchange method [43]. Application of these methods and technologies can reduce the risk of fluoride and arsenic poisoning from water supplies. In addition, local governments and relevant organizations also need to provide public health education to residents, strengthen protection of children, and strengthen the awareness of residents and their knowledge of domestic drinking water treatment.

Conclusions

Fluorine and arsenic concentrations in the rivers of Kyrgyzstan and the number of samples that exceeded the maximum permissible limits for drinking water increased year by year from 2016 to 2018. The fluorine sampling points that exceeded the limit were located in basins or valleys, while the arsenic sampling points that exceeded the limit were located in the Ak-Suu reach of the Chu River Valley. During the observation period, there were no sampling points in Kyrgyzstan at which fluorine and arsenic exceeded the limits simultaneously, but both fluorine and arsenic had high gathering values in the southern Fergana Basin, which indicated that measures should be taken to prevent joint poisoning by fluorine and arsenic in this area. In Kyrgyzstan, the health risk to children was higher than that of adults, which indicated that children's health with respect to river water needs to be protected.

In particular, the carcinogenic risk of arsenic by the ingestion intake route exceeded the maximum acceptable level. Statistical analysis showed that the fluorine and arsenic distributions in the rivers of Kyrgyzstan were mainly affected by mining, industry, agriculture and other anthropogenic activities and that the management and control of fluorine and arsenic in the rivers of Kyrgyzstan will protect the health of local residents from the effects of mining, industry and agriculture.

Methods

Sampling and laboratory analysis.

In this study, samples were taken from the main rivers and their tributaries in Kyrgyzstan from 2016 to 2018. The locations and specific information of the sampling points in each year are shown in Figure 1 (d) and Tables S1-S3. From 2016 to 2018, 83, 92 and 94 samples were collected, with a total of 169 samples. Before sample collection, the containers and samplers were cleaned with detergent, washed with tap water and distilled water and dried in a dry environment. To prevent cross contamination, when sampling at all sampling points, the sampling bottles were moistened with river water three times, and the water samples were then collected. After collection, all water samples were carefully sealed with appropriate labels and stored at 2~5°C for cold storage to inhibit microbial activity and slow down the physical, chemical and biological effects in the water samples; they were transferred to the Central Asia Ecological and Environmental Research Center (Bishkek) of Kyrgyzstan for hydrochemical analysis within 24 hours. The concentrations of the main ions (e.g., HCO_3^-, Cl^-, SO_4^{2-}, NO_3^-, Ca^{2+}, Na^+, Mg^{2+} and K^+) and F^- in the samples were determined by ion chromatography with a Dionex ICs 900 ion chromatography system (Thermo Fisher Scientific Inc., Waltham, Ma, USA). HCO_3^- was determined by the potentiometric titration method, which used a G20 compact titrator (Mettler Toledo AG, Greifensee, Switzerland). Arsenic levels in the river water samples were determined by inductively coupled plasma mass spectrometry using an Agilent 8800 system (Agilent Technologies, Santa Clara, California, USA), with a detection limit of 0.006 μg/L.

Analysis of cold and hot spots

Cold and hot spot analysis is a method to explore the characteristics of local spatial clustering distributions and is used to distinguish and locate hot spots (high value areas) and cold spots (low value areas) in the spatial concentrations of pollutants. The Getis-Ord G^*_i index is often used to describe the spatial distributions of pollutants in cold and hot spot analysis[44], and the calculation formula for G^*_i is as follows:

$$G_i^* = \sum_j^n r_{ij} a_j / \sum_j^n a_j \qquad (1)$$

$$Z = \frac{G_i^* - E\left(G_i^*\right)}{\sqrt{Var\left(G_i^*\right)}} \qquad (2)$$

where G_i^* is the aggregation index of a unit space; Z is the significance of the aggregation index; r_{ij} is the spatial weight; a_j is the attribute value of spatial unit j; $E\left(G_i^*\right)$ and $Var\left(G_i^*\right)$ are the mathematical expectation and variance of G^*_i respectively.

Health risk assessment

In view of the river water use in Kyrgyzstan, the carcinogenic and noncarcinogenic risks of fluorine and arsenic in rivers to adults and children in Kyrgyzstan were evaluated for the drinking water and skin contact routes in this study. Among them, fluorine is a toxic substance (noncarcinogen), and arsenic is not only a noncarcinogen but is also a carcinogen. The parameters of the carcinogenic risk assessment model and noncarcinogenic risk assessment model for different pathways are shown in Table 6.

(1) Health risk assessment for the ingestion intake route

The model [45], which recommended by the United States Environmental Protection Agency (USEPA) was used to evaluate the health risks due to pollutants through drinking water. The carcinogen risk assessment and noncarcinogen risk assessment models are as follows:

$$R_i^c = \left[1 - \exp\left(-D_i \times SF\right)\right] / w \qquad (3)$$

$$R_i^m = \left(D_i \times 10^{-6} / RfD\right) / w \qquad (4)$$

$$D_i = IR \times C_i / BW \qquad (5)$$

where R_i^c is the individual annual average risk due to health hazards caused by carcinogenic pollutants through ingestion intake, a^{-1}; D_i is the average daily exposure dose per unit weight of pollutants through ingestion intake, mg/(kg·d); *SF* is the carcinogenic intensity coefficient of carcinogens entering the human body by ingestion intake, mg/(kg·d); *w* is the average life expectancy, a; R_i^m is the individual annual risk of health hazards caused by noncarcinogenic pollutants by ingestion intake, a^{-1}; *RfD* is the reference daily intake dose of pollutants, mg/(kg·d); *IR* is the average daily ingestion intake volume, L/D; C_i is the mass concentration of pollutants, mg/L; and *BW* is the per capita body weight, kg.

(2) Health risk assessment for the dermal exposure route

The health risk caused by dermal exposure to pollutants was evaluated by the calculation model proposed by Strenge[46], and the specific calculation equation is as follows:

$$R_b^p = \left[1 - \exp\left(-D_b \times SF\right)\right] / w \qquad (6)$$

$$R_b^f = \left(D_b \times 10^{-6} / RfD\right) / w \qquad (7)$$

$$D_b = I_b \times A_{sb} \times FE \times EF \times ED / \left(BW \times AT \times f\right) \qquad (8)$$

$$I_b = 2\times10^{-3}\times k\times C_b\times\sqrt{\frac{6\times\tau\times TE}{\pi}} \quad (9)$$

where R_b^p is the individual average annual risk of health hazards caused by carcinogenic pollutants by dermal exposure, a^{-1}; D_b is the average daily exposure dose per unit body weight of pollutants by skin contact, mg/(kg·d); R_b^f is the average annual risk of health hazards caused by dermal exposure to noncarcinogenic pollutants, a^{-1}; I_b is the adsorption capacity of pollutants by skin for each bath, mg/(cm^2·time); A_{sb} is the body surface area, cm^2; *FE* is the bathing frequency, times/D; *EF* is the exposure frequency, d/a; *ED* is the exposure delay, a; *AT* is the average exposure time, a; *f* is the intestinal adsorption ratio; *k* is the skin adsorption parameter, cm/h; C_b is the mass concentration of pollutants, mg/L; *τ* is the delay time, h; and *TE* is the bath time, h.

(3) General model of water environment health risk assessment

Without considering the synergistic or antagonistic effects of pollutants, the total health risk of the water environment can be expressed as follows [47]:

$$R_{total} = R^c + R^m + R^p + R^f \quad (10)$$

Table 6 Parameters of the health risk assessment model

Parameters	Unit	Parameter reference value		Reference
		Adult	Children	
SF	mg/ (kg·d)	15		48
w	a	70		17
RfD (Arsenic)	mg/(kg·d)	0.0003		49
RfD (Fluorine)	mg/(kg·d)	0.06		
IR	L/d	1.5	0.7	50
BW	kg	60	15	
Asb	cm^2	16000	6660	
FE	times/d	0.3		
EF	d/a	350		51
ED (carcinogen)	a	70		
ED (noncarcinogen)	a	35		

AT (carcinogen)	a	70	
AT (noncarcinogen)	a	35	
f	-	1	
k	cm/h	0.001	
τ	h	1	
TE	h	0.4	[52]

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Figures

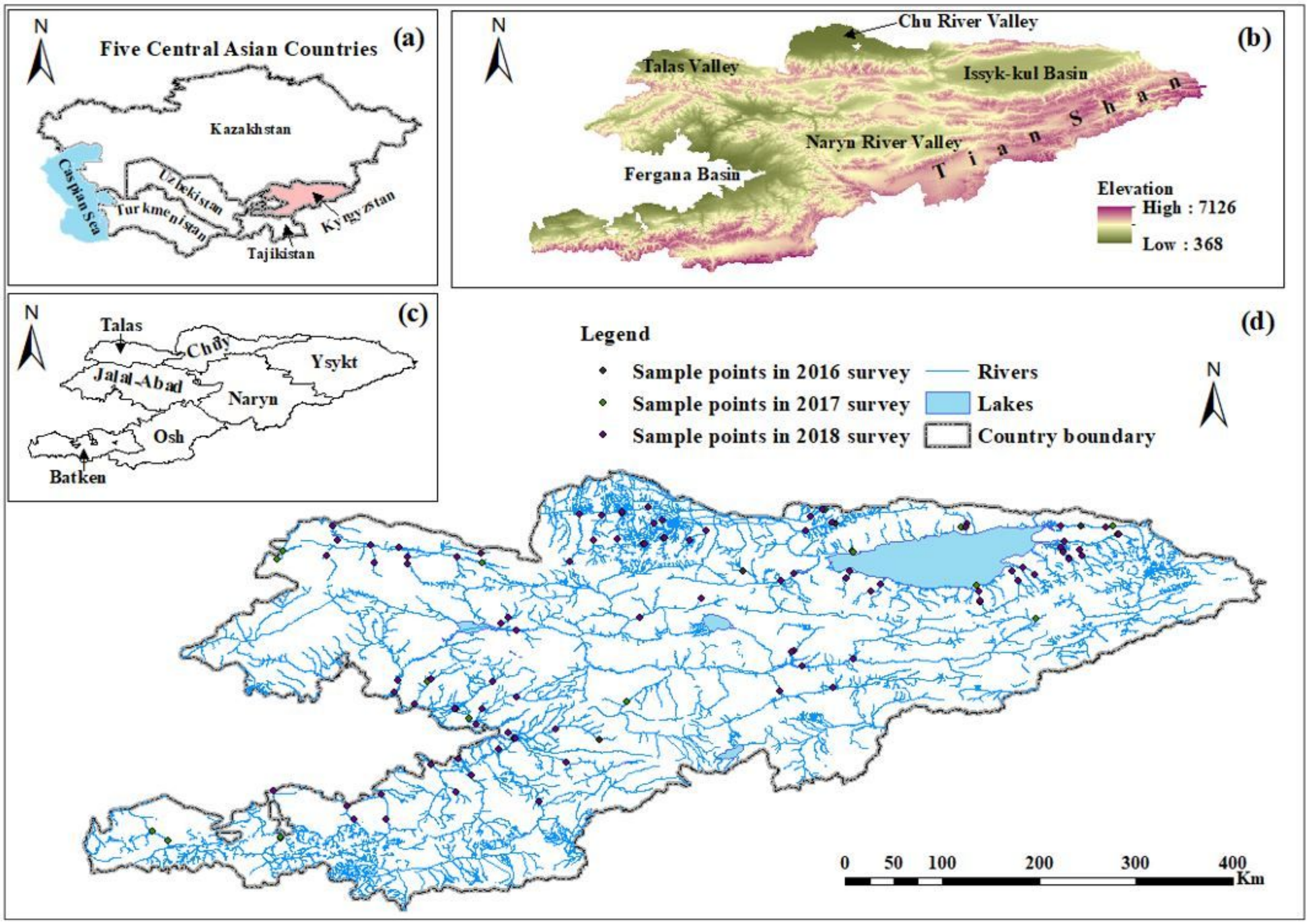

Figure 1

Location of the study area Note: The designations employed and the presentation of the material on this map do not imply the expression of any opinion whatsoever on the part of Research Square concerning the legal status of any country, territory, city or area or of its authorities, or concerning the delimitation of its frontiers or boundaries. This map has been provided by the authors.

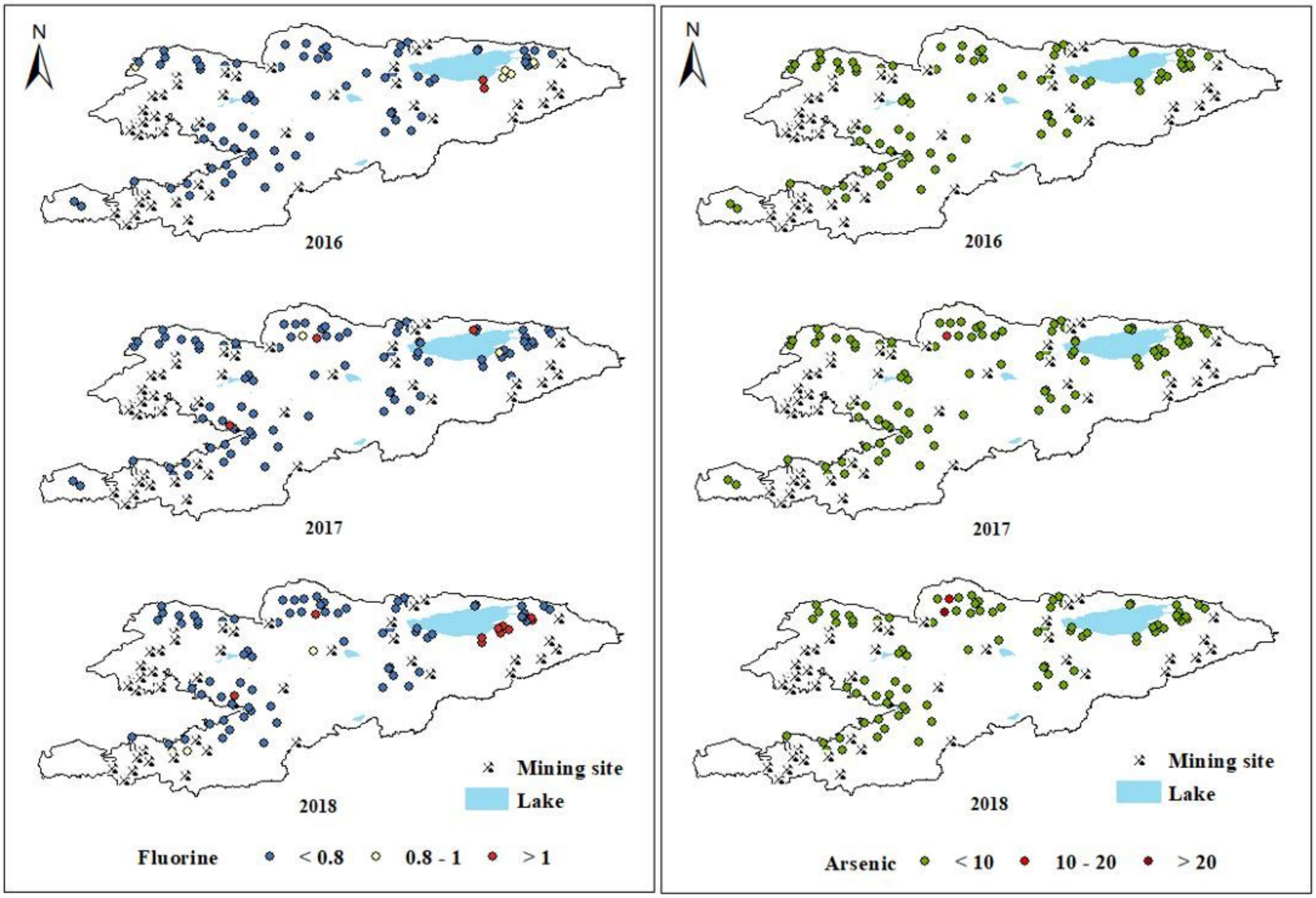

Figure 2

Spatial distributions of fluorine and arsenic in the surface waters of Kyrgyzstan from 2016 to 2018 Note: The designations employed and the presentation of the material on this map do not imply the expression of any opinion whatsoever on the part of Research Square concerning the legal status of any country, territory, city or area or of its authorities, or concerning the delimitation of its frontiers or boundaries. This map has been provided by the authors.

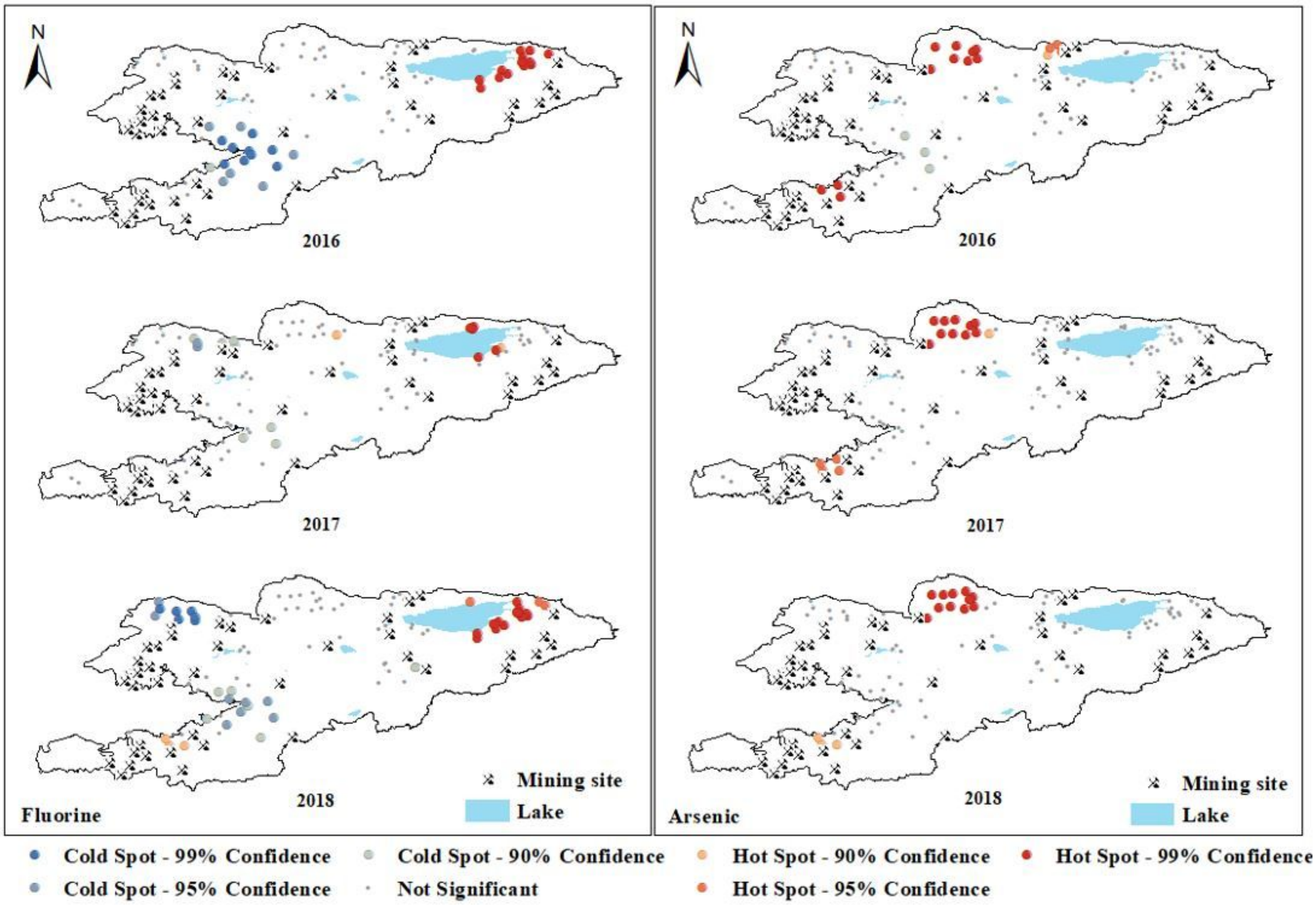

Figure 3

Distributions of cold and hot spots of fluorine and arsenic in the surface waters of Kyrgyzstan from 2016 to 2018 Note: The designations employed and the presentation of the material on this map do not imply the expression of any opinion whatsoever on the part of Research Square concerning the legal status of any country, territory, city or area or of its authorities, or concerning the delimitation of its frontiers or boundaries. This map has been provided by the authors.

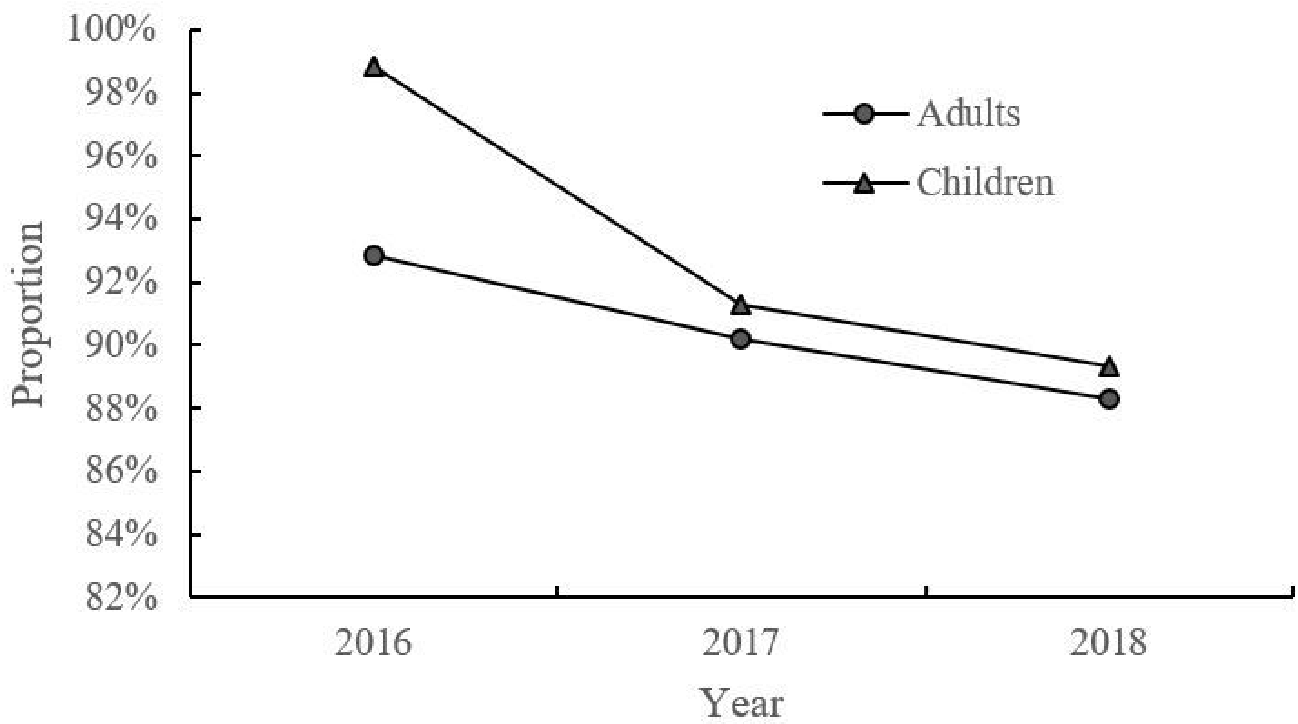

Figure 4

Proportions of carcinogenic risk caused by arsenic through ingestion intake which exceeds the maximum acceptable risk level

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