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ORIGINAL PAPER

Spatial–Temporal Variation and Health Risk Assessment of Fluoride in Surface Water in the Tibetan Plateau

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Abstract

The Tibetan Plateau (TP) is known as the “Asian Water Tower” and provides vital drinking water for residents of China and Southeast Asian countries. However, large-scale regional research on water quality in this climate-sensitive and ecologically-fragile area is still lacking. Considering that drinking from fluoride-contaminated water poses serious health concerns worldwide, especially in Asian counties, it is urgent to clarify the spatial–temporal distribution characteristics, influencing factors, and health risk of fluoride in surface water in the TP. In this study, a total of 2697 surface water samples from major rivers and typical lakes in the TP were systematically analysed. Overall, fluoride concentrations ranged from 0.003 to 6.240 mg L^{-1} and varied among water periods, water basins and even water types. Pearson’s correlation analysis showed that the distribution of fluoride concentration was closely related to the regional climate and positively correlated with anthropogenic activities. Probabilistic health risk assessment revealed that potential hazards in the Inner Basin were the highest for all age groups ($HR > 1$), especially for infants and adults ($HR > 3$), while the risks in most other water basins were acceptable ($HR < 1$). Our findings can provide scientific support for fluorosis prevention, and guide water resource utilization in the TP and adjacent regions.

Keywords Fluoride · Surface water · Tibetan Plateau · Health risk assessment · Influencing factors

Introduction

Fluoride is widely distributed in the natural environment and closely affects human health (Li et al. 2019; Liu et al. 2021b). In general, 80–90% of the fluoride in the human body is concentrated in the bones and teeth. Appropriate absorption of fluoride is beneficial to human growth and bone metabolism (Meng et al. 2021). However, excessive fluoride intake leads to dental fluorosis, skeletal fluorosis and other diseases, which seriously affect health (Yin et al. 2021a). Numerous studies have shown that high fluoride concentrations in drinking water (> 1.0 mg L^{-1}) can cause dental fluorosis and even bone fluorosis (Ahada and Suthar 2019; Narsimha and Rajitha 2018).

Drinking water with a high fluoride concentration remains a challenge impacting the health of millions of people worldwide (Narsimha and Rajitha 2018). Drinking water fluorosis occurs widely worldwide and is prevalent to varying degrees in more than 50 countries and regions in Asia (India, Bangladesh, China, Thailand, Sri Lanka, etc.), Europe (Russia, Bulgaria, Italy, etc.), Africa, the Americas and Oceania (Ahada and Suthar 2019; Li et al. 2020a; Zhang et al. 2017). China used to have one of the most serious epidemics of drinking-type fluorosis, with 1115 counties, 75,287 villages and 72.07 million people affected (Zhang et al. 2020; He et al. 2020).

The Tibetan Plateau (TP) is one of the areas where both brick tea-type fluorosis and drinking-type fluorosis are prevalent (Lou et al. 2021). Researchers considered drinking brick tea to be the main cause of fluorosis in the TP because the average fluoride content of brick tea in this

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area is approximately 800 mg kg^{-1}, which is far above the national safe threshold for brick tea fluoride concentration (300 mg kg^{-1}) (Zhang et al. 2019b; NHCC 2005). Therefore, previous studies on fluorosis in the TP mainly focused on brick tea, ignoring the endemicity of drinking-type fluorosis, which may mislead fluorosis prevention and treatment in the TP. In addition, it has been confirmed that compared with other fluorosis endemic areas, the same quantity of brick tea fluoride intake in the TP causes more severe fluorosis symptoms (Liu et al. 2020), which suggests that the superposition of fluoride intake via other pathways, such as through drinking water, makes fluorosis more serious. Unfortunately, to date, no data on the water fluoride concentration in the TP are available in the published literature.

The TP is the cradle of major Chinese river systems, including the Yellow River, Yangtze River and Mekong River. Moreover, several important rivers in Southeast Asia, such as the Ganges, Indus and Salween rivers, originate in this region. The TP is considered the “Asian Water Tower”, providing fresh water for nearly one-third of the world’s population (Qiao et al. 2021) and playing a vital role in maintaining human health and economic and social development. The water quality of the TP has a profound influence not only on China but also on all of Southeast Asia. Therefore, it is of great significance to study the distribution and influencing factors of fluoride in water in the TP.

Currently, the distribution of surface water fluoride is thought to be related to both natural and anthropological factors. Hydrochemical characteristics are influenced by regional climate, topography and anthropogenic activities (Liu et al. 2021b; Ma et al. 2021; Zhang et al. 2019b). The temporal and spatial distribution of water resources and changes in water quality caused by climate conditions have generated much research interest from scientists and governments in various countries (Zhang et al. 2021). However, most studies have focused on the impact of climate change on the amount of water, and relatively few studies have concentrated on changes in water quality (Ran et al. 2021; Duan and Duan 2020). There has been almost no discussion on the impact of climate on fluoride distribution in water. Climate change affects the regional surface water cycle by changing the temporal and spatial distribution of meteorological factors such as air temperature and precipitation, influencing the migration and transformation of compounds and ions in the water (Wu 2016; Duan and Duan 2020; Nury et al. 2021; Yin et al. 2021b). Moreover, the variability in topography in the plateau region leads to an uneven distribution of surface elements and makes the region prone to endemic diseases (Xu et al. 2018; Hettithanthri et al. 2021). Anthropogenic activities, such as effluent dischange, and agriculture, also influence hydrochemical characteristics (Zhang et al. 2019b; Ma et al. 2021).

Therefore, the purposes of this study are: (1) to clarify the spatial–temporal variation of fluoride concentrations in the surface water of major rivers and typical lakes in the TP, (2) to estimate the health risks posed by fluoride and (3) to assess the possible factors affecting the concentration and distribution of fluoride in the TP. This study will be helpful to decision makers with a scientific basis of surface water fluoride for fluorosis prevention in the plateau region.

Materials and Methods

Study Area

The study area includes the Qinghai Province and the Tibet Autonomous Region (78°23′24″–103°4′12″ E; 26°51'–39°12′36″ N) comprising the majority of the area of the TP (60.6%). The region covers an area of 1.9507 million square kilometres and 118 counties with a total population of 9.43 million (Fig. 1). The TP is the highest of “the three steps of China's terrain”, with an average elevation of 4400 m (Wang et al. 2017). Several famous rivers originate here, including the Yangtze, Yellow River, Brahmaputra, Salween and Indus rivers. The area also has a high density of lakes, and the lakes in the TP account for 57% of the total lake area and 69% of the inland lake area in China (Pi et al. 2020; Lu et al. 2020). The annual average air temperature of the TP ranges from − 6 °C in the northwest to 20 °C in the southeast (Dong et al. 2020). The annual precipitation is uneven, decreasing from the southwest (2000 mm) to the northwest (less than 50 mm) (Wu et al. 2019).

The main sources of surface water in the TP are precipitation and melting ice, with a small interannual variation in river flow (Nury et al. 2021; Wang et al. 2020b). In general, the annual flow is mainly concentrated from June to October (rich period), accounting for 75 to 80% of the annual main flow, and the dry period ranges from December to April, while the remaining months represent flat periods (Liu et al. 2021c). It is worth mentioning that the water periods in this study were determined by actual measurements in each month, and the specific time of the three periods may vary slightly according to different natural factors, such as temperature, precipitation or vegetation cover (Xu et al. 2021).

Data Sources

Water Quality Data

Water quality data in this study were derived from the China National Eco-environmental Monitoring Network (http://www.cnemc.cn/). The dataset contained a total of 2697 samples which were collected monthly from 69 monitoring sampling sites (61 river sampling sites and 8 lake sampling

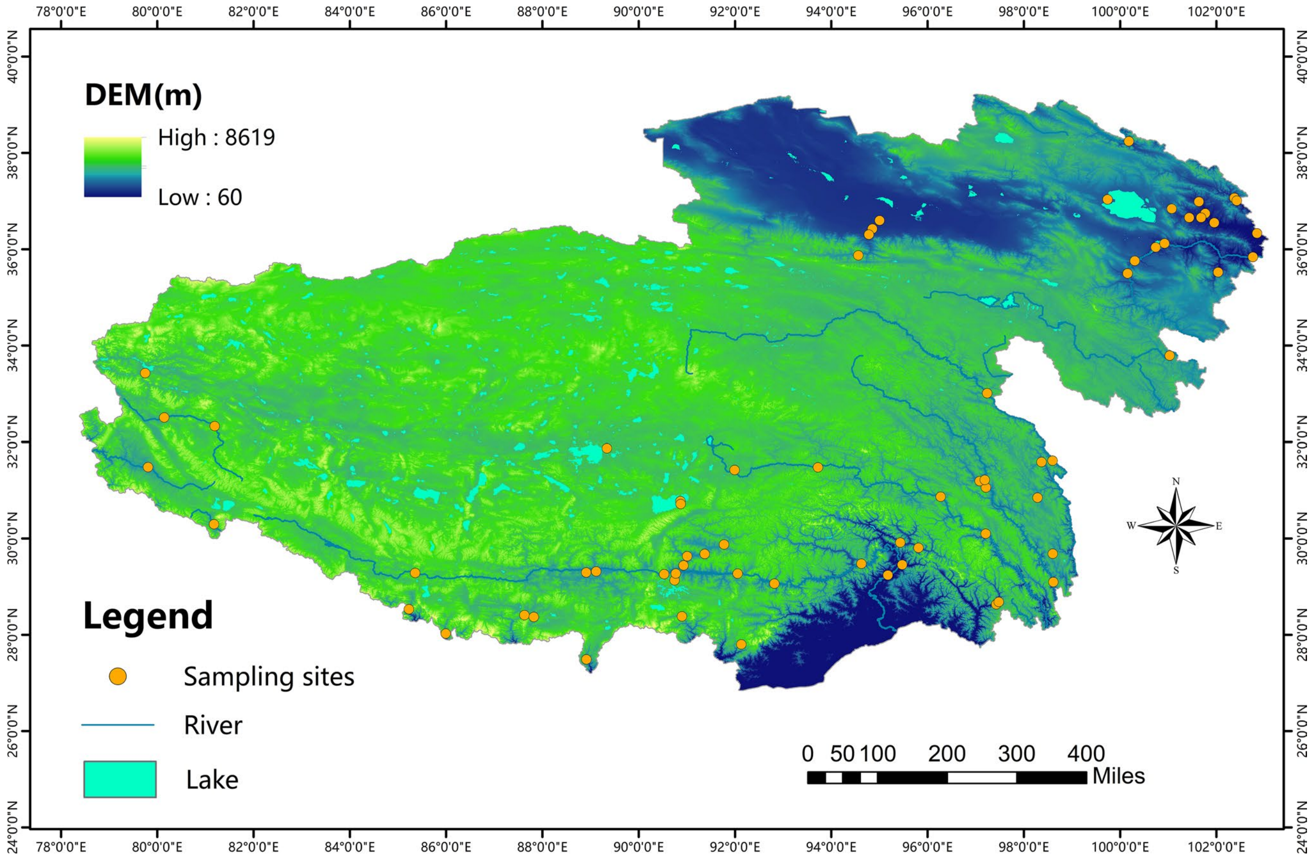

Fig. 1 Spatial distribution of national surface water monitoring stations in the TP

sites) throughout the TP, covering a full 5-year (2016–2020) period.

Water samples were collected by using a hydrophore, stored in polyethylene bottles, and acidified with high-purity concentrated HNO_3 to a pH less than 2. Ion chromatography was used to determine the fluoride concentration. In addition, there was a series of standard quality control process, including full-programme blank sample, parallel samples, black test, correlation test, continuous calibration, precision control and accuracy control. Specific steps about sampling collection, laboratory analysis, and quality control methods were added to Supplementary Files.

Watershed Landscape Data

To obtain the water basin dataset for the TP, we used the dataset of TP river basin maps from the National Tibetan Plateau Data Center (TPDC) (Zhang et al. 2019a). The study area was divided into 10 water basins (Brahmaputra, Ganges, Hexi, Indus, Inner, Mekong, Qaidam, Salween, Yangtze and Yellow River) according to the river network and elevation (Fig. 2). Elevation was obtained from the Resource and Environment Science and Data Center (https://www.resdc.cn/). Slope data were transformed from elevation by using the ArcGIS Surface Toolset.

Sampling sites were set according to the population and water basins. Therefore, the sampling sites were unevenly distributed among the water basins. For example, 32% of the sampling sites were in the Brahmaputra Basin, compared with barely 3% in the Inner Basin (Fig. 2). Therefore, we divided the water basins into three categories according to the location of the sampling sites and the distance between sampling sites, the main river stream and the population areas: (1) Uniform type, including the Brahmaputra, Ganges, Indus, Salween, Yangtze and Yellow River Basins, in which sampling sites covered the main stream and densely populated areas and represented the whole water basin well. (2) Scatter type, containing the Hexi, Mekong, and Qaidam Basins, in which sampling sites were few and scattered and did not fully cover the densely populated areas. (3) Inner type, a separate classification of the Inner Basin according to its inner lake density, river scarcity, and underpopulation.

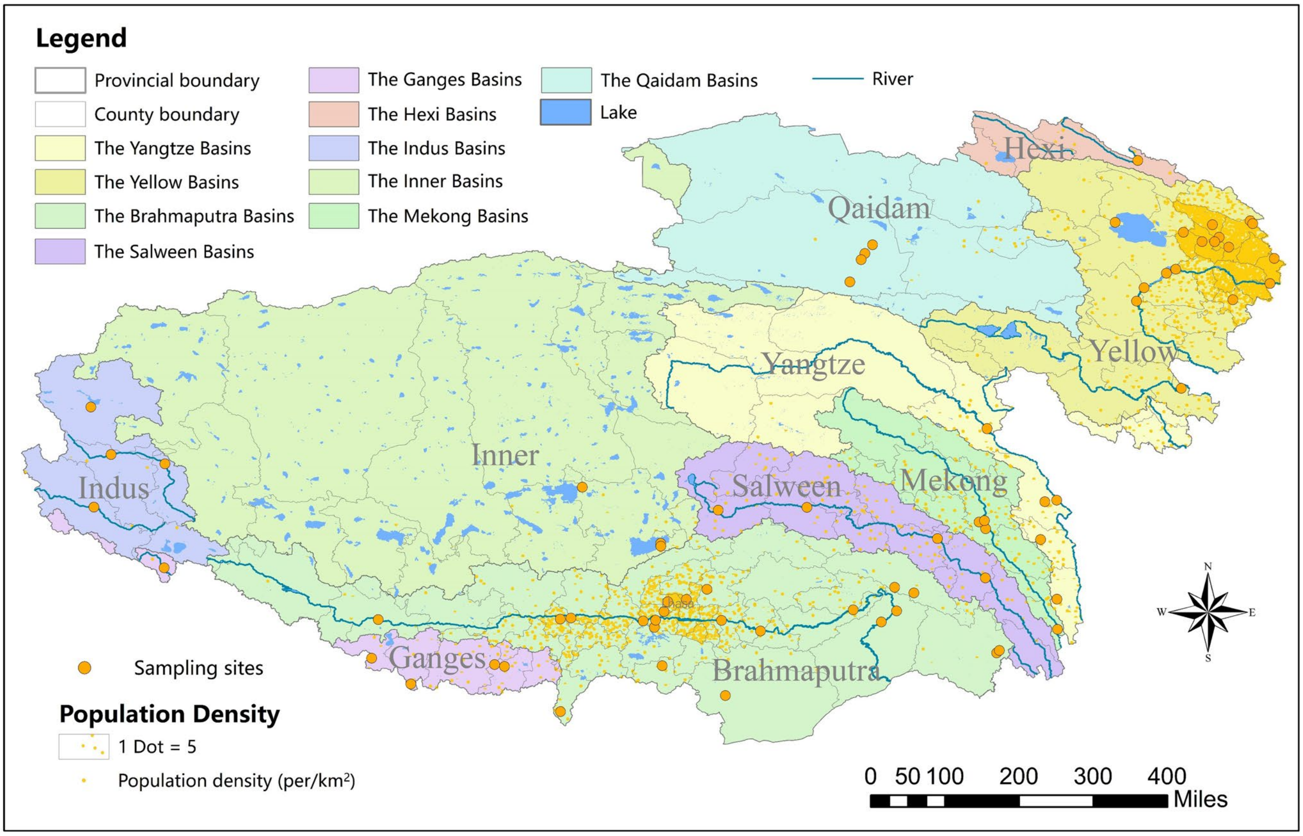

Fig. 2 Water basins and population density

Meteorological Data

Local meteorological data for the TP, including air temperature (°C) and precipitation (mm d^{-1}) data, were obtained from the Chinese Meteorological Forcing Dataset provided by the TPDC (Yang ad He 2019). The dataset has a high spatial resolution of 0.1°, with gridded near-surface meteorological data developed specifically for studies of land-surface processes across the TP. The original data of our precipitation dataset came from meteorological bureau observations, reanalysis data and satellite remote sensing data.

Night Light Index

In this study, night light data obtained from the visible light imaging linear scanning system of the U.S. National Polar Orbiting Satellite Visible Infrared Imaging Radiometer (VIIRS) (https://earthdata.nasa.gov/). The spatial resolution of the VIIRS is 0.75 km, and the spatial resolution of the night light products is 0.5 km.

Night light index as a data source for retrieving the intensity of anthropogenic activities has been widely used in the field of environmental sciences (Xu and Gao 2017). A large number of existing studies have shown that the night light index is positively correlated with the economic development level, energy consumption level and population density of the region (Fu et al. 2018; Kang and Jung 2019; You et al. 2021).

Statistical Analysis

ArcGIS 10.4 software was used to present the spatial distribution of the surface water fluoride concentration. Statistical analysis was conducted in SPSS software (IBM SPSS Statistics 26), and Pearson's rank correlation coefficient was constructed to analyse the relationship between fluoride and influencing factors. In addition, the human health risk of surface water fluoride in the study area was assessed based on the health risk assessment model recommended by the United States Environmental Protection Agency (USEPA) (USEPA 1989), and the sensitivity analysis was applied with the Monte Carlo method using Crystal Ball software (Oracle Crystal Ball version 11.1.2.3).

Human Risk Assessment

In this study, the potential human health risk of fluoride was assessed for different age groups because of their different behavioural and physiological attributes (USEPA 2008). According to the differences in environmental exposure behaviour among different age groups, we divided the potentially affected population into five groups: infants (0–3 years old), young children (4–6 years old), children (7–12 years old), teenagers (13–18 years old) and adults (> 18 years) (Wang et al. 2018). Fluoride in water acts as a toxic but noncarcinogenic substance (Wang and Li 2022). The risk assessment of a toxic substance is based on the reference dose (Li et al. 2020a).

Fluoride in water enters the human body through ingestion, dermal absorption and inhalation. However, inhalation was not included in this study. Some toxicological data, such as the inhalation reference dose for fluoride and the transfer efficiency from water to air, are unavailable in the database and published literature (Agency for Toxic Substances and Disease Registry 2003; Zhang et al. 2020). Considering the two other exposure pathways, we estimated the daily exposure dose of fluoride through water ingestion (oral intake of surface water) and dermal absorption (dermal contact with surface water through daily bathing) (Li et al. 2020a; Zhang et al. 2020). The formulas for calculating the average daily exposure doses I_i and I_d are as follows, and the values of the parameters are shown in Table 1.

$$I_i = \frac{\mathrm{C} \times \mathrm{IR} \times \mathrm{EF} \times \mathrm{ED}}{\mathrm{BW} \times \mathrm{AT}}, \tag{1}$$

$$I_d = \frac{\mathrm{C} \times \mathrm{SA} \times K_{\mathrm{p}} \times \mathrm{F} \times \mathrm{ETs} \times \mathrm{EF} \times \mathrm{ED} \times 10^{-3}}{\mathrm{BW} \times \mathrm{AT}}, \tag{2}$$

where I_i represents the estimated average daily exposure dose of fluoride through ingestion (mg kg^{-1} d^{-1}); I_d represents the estimated average daily exposure dose of fluoride through dermal absorption (mg kg^{-1} d^{-1}); C is the measured concentration of fluoride in water (mg L^{-1}); IR is the drinking rate (L d^{-1}); EF is the exposure frequency (d a^{-1}); ED is the exposure duration (a); BW is the average weight (kg). AT is the life expectancies of residents (d); SA is the average skin surface area (m^2); K_p is the dermal permeability constant: 10^{-3} (cm h^{-1}); F is the fraction of surface skin contact with water: 0.4 (unit-less); ETs is the exposure time (h day^{-1}); 10^{-3} is the number of L per cm^{-3}.

The noncarcinogenic risk assessment model used in the present study was as follows:

$$\mathrm{HR} = \frac{\sum \mathrm{I}}{D_{\mathrm{Rf}}} \tag{3}$$

where HR is the noncarcinogenic risk index and D_{Rf} represents the reference dose of a noncarcinogenic substance (mg kg^{-1} d^{-1}). The fluoride reference dose for human health risk by exposure through consumption of potable water is 0.06 mg kg^{-1} d^{-1}.

The USEPA health risk assessment guidelines indicate that the threshold of noncarcinogenic risk HR is 1 (USEPA 1989). An HR value < 1 indicates that the health risk posed by noncarcinogenic substances is within an acceptable level, whereas an HR > 1 indicates that the noncarcinogenic health risk is unacceptable.

All data sources in this study are shown in Supplementary Table 1.

Results and Discussion

Spatial–Temporal Variation in Surface Water Fluoride in the Tibetan Plateau

The overall statistical characteristics of fluoride in surface water in the TP are shown in Table 2. In total, 2697 samples collected monthly over a period spanning 2016–2020 were analysed. From 2016 to 2020, the average fluoride concentration showed a downward trend, decreasing from 0.408 to 0.278 mg L^{-1}. In general, the median concentration was approximately 0.23 mg L^{-1}, and the standard deviation was approximately 0.68. The concentration varied greatly among sampling sites. The maximum concentration exceeded 4.0 mg L^{-1} each year, while the minimum value stayed at approximately 0.005 mg L^{-1}.

Table 3 is a statistical summary of the fluoride content of surface water in the TP among the dry, flat and rich periods. The fluoride concentration during the dry periods ranged from 0.003 to 4.560 mg L^{-1}, with a 5-year average value of 0.292 mg L^{-1}. The 5-year average concentrations in the flat and rich periods were 0.437 and 0.376 mg L^{-1}, respectively. The highest concentration came from Namco every year from 2016 to 2020, exceeding the fluoride limit of 1 mg L^{-1} in the "Standards for drinking water quality" (NHCC 2006) and the fluoride limit of 1.5 mg L^{-1} in the "Standards for surface water quality" (NHCC 2002) of China.

Spatial Differentiation of Surface Water Fluoride in the Tibetan Plateau

Our results showed that the TP surface fluoride concentration displayed spatial variation among the 10 water basins. The mean concentrations in the water basins were in the following order: Inner > 1 mg L^{-1} > Qaidam > Mekong > Salween > Yellow River > Indus > Yangtze > Brahmaputra > Hexi > Ganges.

As shown in Fig. 3, the surface water fluoride in the Uniform-type region was lower than 0.50 mg L^{-1}. However, various spatial distributions were observed among water basins. The fluoride concentration in the Salween Basin remained evenly distributed at approximately 0.30 mg L^{-1}. The Brahmaputra flows from west to east, with relatively

Table 1 Human exposure parameters of fluoride (Duan 2012a, b)

Parameters	Infants (0–3)	Young children (4–6)	Children (7–12)	Teenagers (12–18)	Adults (> 18)
IR	0.49	0.78	1.09	1.59	2.71
EF	365	365	365	365	365
ED	1.5	5.0	9.5	15.0	52.0
BW	9.21	16.65	28.15	47.3	58.55
AT	548	1825	3468	5475	18,980
SA	0.45	0.72	1.04	1.47	1.60
ETs	0.069	0.078	0.100	0.120	0.083

high concentrations in the Lhasa river, the biggest tributary of the Brahmaputra (approximately 0.40 mg L^{-1}). The southwestern region of the TP, where the Indus and Ganges Basins are located, mainly had low values (< 0.25 mg L^{-1}). The fluoride concentration of two important water basins in China, the Yellow River and Yangtze River, were also below the fluoride permissible limit of China national standards for drinking water quality (NHCC 2006).

In the Scatter-type regions, sampling sites were concentrated in the middle and lower reaches of the Mekong Basin in our study area, which had concentrations ranging from 0.25 to 0.50 mg L^{-1}. The smallest basin in this study was Hexi. Although there was only one sampling site there, it maintained quite a low value (< 0.25 mg L^{-1}). The landscape of the Qaidam Basin is an arid desert, and the waters there are mainly endorheic lakes and their inflows. According to our research, the fluoride concentration in the south-central Qaidam Basin ranged from 0.25 to 1.00 mg L^{-1}.

Extremely high values appeared in the Inner-type region, where surface water was sampled from two typical endorheic lakes (Namco and Siling Co). Figure 3 shows the water fluoride concentration in the Namco. Consistent with a study by Wang et al. (2010), the concentration of the major ions in the Namco's water was higher than that in the stream water.

The fluoride concentration in lakes was higher than that in rivers in the TP in this study, with average values of 1.36 and 0.29 mg L^{-1}, respectively. Five typical lakes (Yamzhog Yumco, Longyangxia Reservoir, Bangong Co, Namco and Siling Co) in this study were taken into consideration to explore the large differences between lake and river concentrations. We analysed rivers, lakes and lake-inflows in the same water basin according to their flow direction and confluence area. Figure 4 clearly shows that fluoride concentrations in lakes were higher than those in rivers and lake-inflows in the same water basin. Studies related to fluoride in surface water in the TP published from 2010 to 2021 shown similar trends (Supplementary Table 2).

The fluoride concentrations in the Yamzhog Yumco and Longyangxia Reservoirs, with average values of 0.64 mg L^{-1} and 0.35 mg L^{-1}, respectively, were 2–3 times higher than the fluoride concentrations in their inflows. Moreover, the average values of rivers in the Brahmaputra, Yellow River and Indus Basins remained at approximately 0.20 mg L^{-1}, while the mean (4.11 mg L^{-1}) and median (4.28 mg L^{-1}) fluoride concentrations in Namco were considerably high. Compared to rivers, the other typical lake (Siling Co) in the Inner Basin also had a considerably high average concentration (0.61 mg L^{-1}).

There may be several influencing factors according to the literature. Evaporation plays a certain role in the increase in ions in lake water (Wang et al. 2020a). The lakes in the TP are mainly endorheic; thus, fluoride is concentrated in the lakes as water evaporates. It has also been reported that the water ion composition of Namco is dominated by evaporation and crystallization (Ma et al. 2016). The geological processes of confluence and rock weathering affect

Table 2 Overall statistics of fluoride concentration in surface water in the TP from 2016 to 2020

Year	Average (mg L^{-1})	Max (mg L^{-1})	Min (mg L^{-1})	Standard deviation	Median (mg L^{-1})	Sample sizes (N)
2016	0.408	4.730	0.010	0.663	0.238	623
2017	0.359	4.830	0.003	0.653	0.236	814
2018	0.368	6.080	0.003	0.738	0.231	544
2019	0.333	6.240	0.006	0.777	0.245	446
2020	0.278	4.091	0.006	0.597	0.179	270

Table 3 Fluoride concentration in dry, flat and rich periods in the TP

Parameters	5-year average (mg L^{-1})	Max (mg L^{-1})	Min (mg L^{-1})	Median (mg L^{-1})	Standard deviation	Sample sizes (N)
Dry period	0.381	4.560	0.003	0.227	0.612	1079
Flat Period	0.366	6.240	0.009	0.227	0.621	777
Rich period	0.395	6.080	0.013	0.226	0.728	841

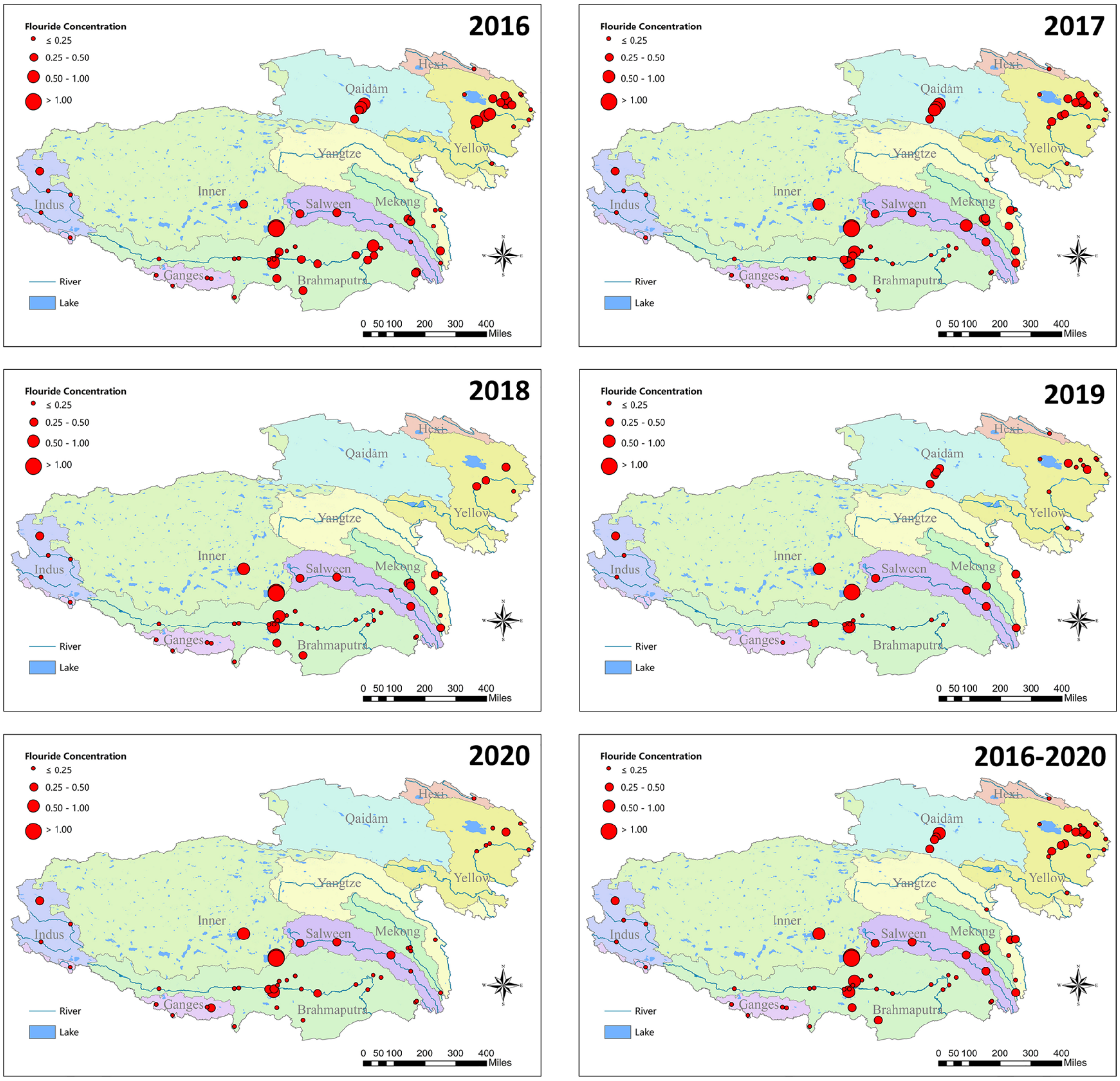

Fig. 3 Spatial patterns of annual average and 5-year average water fluoride in the TP during 2016–2020

river water ion concentrations (Yu et al. 2021). Additionally, weathering is a main factor causing the increase in all ions in the lake water and most ions in river water (Wu 2016). According to the existing research on water in the TP, the chemical composition changes after a river flows into a lake, and the main positive ions changed from ($Ca^+ + Mg^+$) to ($Na^+ + Mg^+$) (Wang et al. 2010). Water F^- was found to be positively correlated with Na^+ and presented a significantly negative relationship with Ca^{2+} (Liu et al. 2021a). A series of complex chemical processes occurred in the lake water, accumulating fluoride to some extent.

Temporal Differentiation of Surface Water Fluoride in the Tibetan Plateau

Regarding the changes in fluoride concentration over time, our analysis showed that the fluoride concentrations in the three different periods (dry period, flat period and rich period) remained relatively stable, changing little in general. However, there were some variabilities. The fluoride concentration in the whole Ganges Basin showed a higher value (0.25 to 0.50 mg L^{-1}) in the flat period of 2018 than

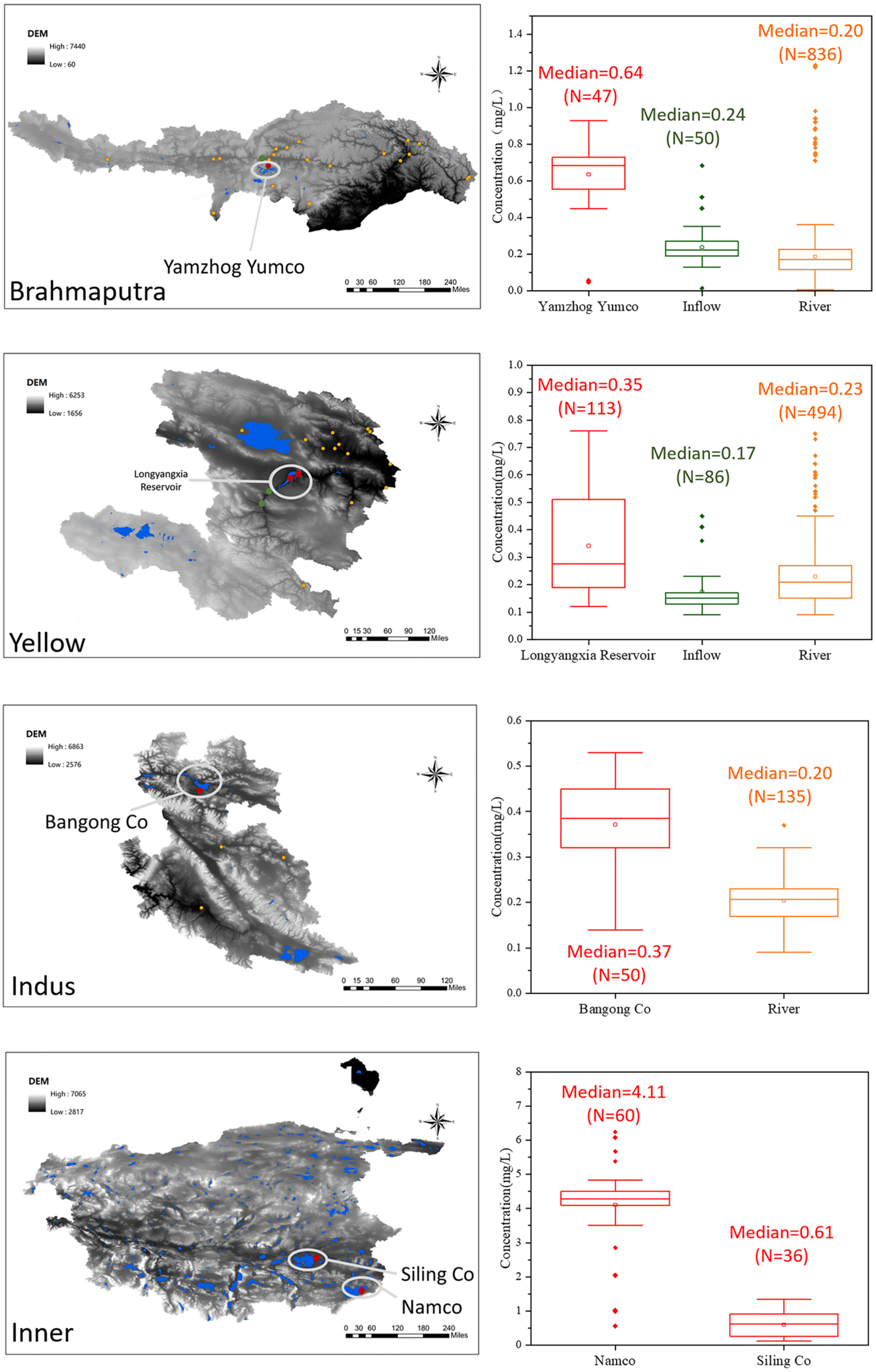

Fig. 4 Sampling distribution and boxplot of fluoride in different water basins among rivers, lakes and inflows

in other periods during 2016–2020. In areas with a high population density, such as the central of Brahmaputra Basin, the northeastern of Yellow River Basin and the middle of Mekong Basin, the fluoride concentration in the three periods of 2020 presented a lower value than that in the same periods during 2016–2019. This may have been due to decreases in anthropogenic activities in 2020 for COVID-19, illustrating that anthropogenic activities may affect surface water fluoride.

In addition, there was a downward tendency in the fluoride concentration each year. For instance, Fig. 5 shows that fluoride concentrations in the Yellow River Basin

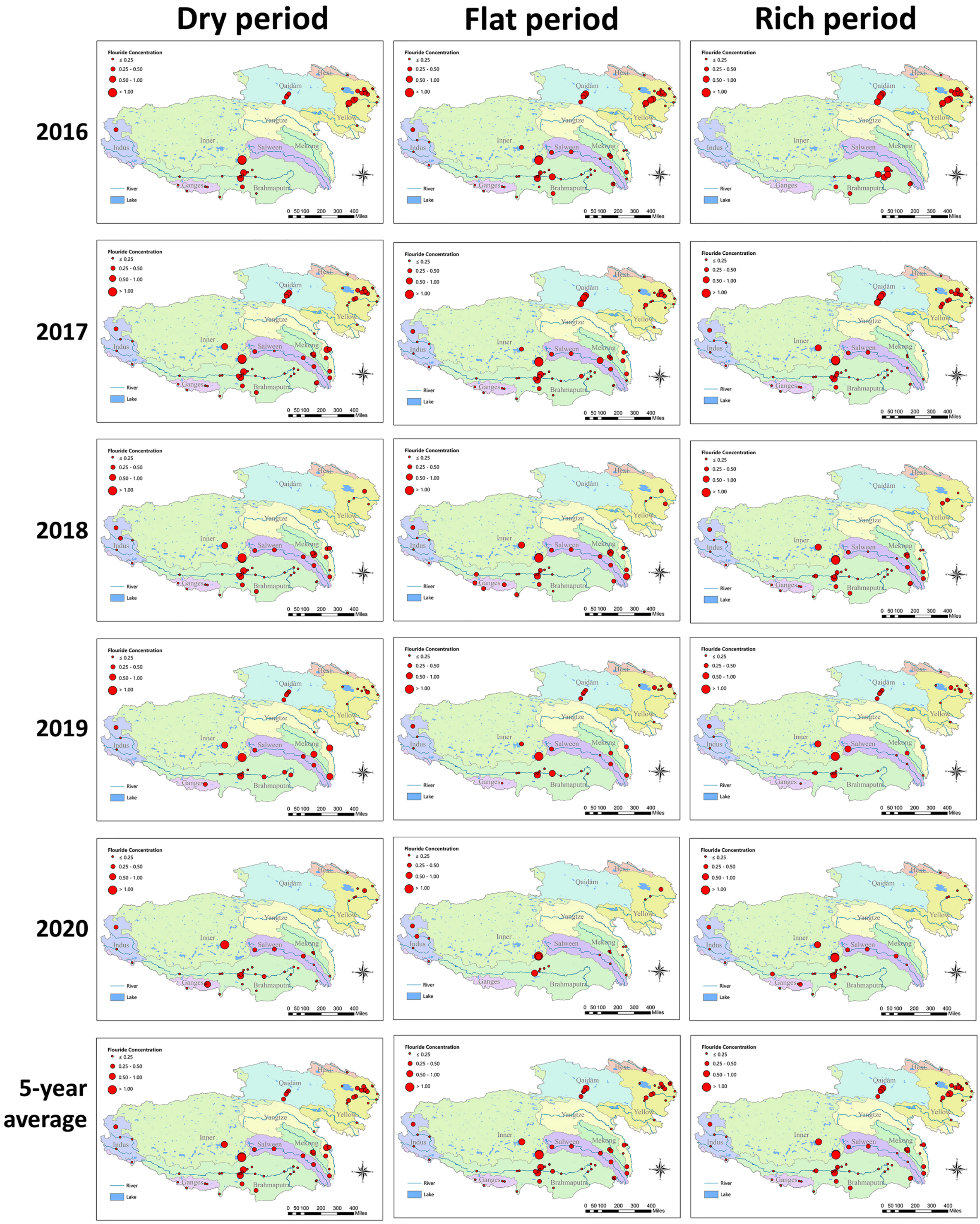

Fig. 5 Variability in water fluoride in dry, flat, and rich periods during 2016–2020

during all three water periods had an obvious declining trend. The lake fluoride value showed the same decrease, which may be related to the increase in lake water storage for decades caused by climate change—the increasing trend of air temperature and precipitation in the Tibet Plateau from 1975 to 2016 (Yan et al. 2018). Other researchers have also revealed that major ions decreased in response to regional climatic change in Tibetan lakes (Qiao et al. 2021; Yan et al. 2018).

Effects of Anthropogenic Factors and Natural Characteristics on Surface Water Fluoride

To identify the influence of anthropogenic, climate condition and topographical factors on surface water fluoride (Fig. 6), Pearson's rank correlation analysis was applied to the surface water fluoride content in 8 different basins (Brahmaputra, Ganges, Indus, Mekong, Qaidam, Salween, Yangtze and Yellow River) and 5 key factors (night light intensity, air temperature, elevation, precipitation, and slope), while the Hexi and Inner Basins were eliminated due to few sampling sites (Table 4).

The night light index, which is used to measure the intensity of anthropogenic activities, has been widely used in the field of environmental sciences (Zheng et al. 2022). Therefore, the night light index was used to represent the influence of anthropogenic activities in our study. The contrast between bright and dark areas in night light images represents a powerful tool to in the study of intensive anthropogenic activities and their effects (Stathakis and Baltas 2018). Night light intensity showed a positive relationship with fluoride concentration in high population density areas (Brahmaputra, Mekong, Salween, and Yellow River Basins). In particular, the correlations in the Brahmaputra and Salween Basins were 0.477 and 0.972, respectively ($P < 0.05$). The results suggest that anthropogenic activities may increase the water fluoride concentration in high population density areas on the TP.

Concerning the regional climate, the surface water fluoride concentration had a weakly negative relationship with precipitation, a nonsignificant positive relationship with elevation, and a negative correlation with air temperature ($r = -0.296$, $P < 0.05$). Precipitation was negatively correlated with fluoride in nearly all the water basins. During precipitation, water inflows via surface and subsurface processes from soil and thus may significantly modulate water quality and ion concentration (Wu et al. 2019). Therefore, precipitation intensity and frequency directly affect water quality. Air temperature was positively correlated with fluoride in the northeastern area but negatively correlated in the remaining areas. Water temperature changes with air temperature changes. Generally, air temperature can affect the density, surface tension, viscosity and morphology of water bodies and change the distribution of the water temperature layer, the rate of chemical reaction and biodegradation in water bodies, which fundamentally controls the physical and chemical properties and biological characteristics of the water bodies (Giri 2021).

Among numerous topographical factors, surface elevation and slope were the most essential and were recognized as the key factors (Xu et al. 2018). For the relationship between elevation and fluoride, the southeastern TP, including the Mekong ($r = 0.646$), Salween ($r = 0.301$), and Yangtze ($r = 0.955$, $P < 0.05$) Basins, presented a significantly positive relationship. These three basins are close to the first and second steps of China's terrain, with rapid decreases in elevation and efficient water confluence resulting in a diluted fluoride concentration. However, the northeastern TP (Yellow River and Qaidam Basins) revealed a negative but nonsignificant correlation. The slopes in these water basins showed negative but nonsignificant correlations.

Table 4 Pearson's rank correlation coefficients of anthropogenic and natural drivers of fluoride in nine major river basins in TP

Correlation	Night light intensity	Air temperature	Precipitation	Elevation	Slope
All basins	0.043	− 0.296*	− 0.158	0.172	0.003
Brahmaputra	0.477*	− 0.323	− 0.241	− 0.178	0.175
Ganges	− 0.716	− 0.116	− 0.214	0.077	− 0.479
Indus	− 0.452	− 0.241	0.169	− 0.132	− 0.341
Mekong	0.386	− 0.118	− 0.216	0.646	− 0.226
Qaidam	− 0.124	0.769	− 0.729	− 0.791	− 0.337
Salween	0.972*	− 0.557	− 0.673	0.301	0.278
Yangtze	− 0.103	− 0.571	− 0.443	0.955*	− 0.826
Yellow River	0.255	0.225	− 0.540*	− 0.312	0.225

*$P < 0.05$

Table 5 HR values for the five age groups

		Average	Max	Min
Ingesting health risk	Infant (0–3 years)	0.37	4.44	0.11
	Young children (4–6 years)	0.29	3.18	0.10
	Children (7–12 years)	0.26	3.13	0.08
	Teenager (13–18 years)	0.22	2.46	0.07
	Adults (> 18 years)	0.36	4.61	0.11
Dermal health risk ($\times 10^{-4}$)	Infant (0–3 years)	0.51	5.88	0.18
	Young children (4–6 years)	0.76	8.70	0.26
	Children (7–12 years)	0.60	6.84	0.21
	Teenager (13–18 years)	1.33	15.22	0.46
	Adults (> 18 years)	1.08	12.39	0.37

Health Risk of Fluoride in Surface Water in the Tibetan Plateau

Health Risk Assessment

The health risk of fluoride varied by exposure pathway and age groups (Table 5). The ingestion health risk ranged from 0.07 to 4.61, while the health risk via the dermal absorption ranged from 0.18×10^{-4} to 15.22×10^{-4}. This suggests that ingesting is the main exposure pathway causing health risk, and the health risk via the dermal absorption pathway is so small that can be ignored in our study. From an age group perspective, adults and infants faced more ingesting health risks, while the dermal risk faced by teenagers was significantly higher than that for other age groups. In addition, the average health risk for all age groups were less than 1, indicating that the fluoride health risk of surface water in the TP was acceptable on a large scale. However, fluoride will accumulate in human body, and water fluoride intake can exacerbate the existing serious fluorosis in the TP (Fig. 6).

Figure 7 presents the fluoride health risk distribution of TP water basins by age group. The lowest health risk was associated with nearly all age groups in the Hexi Basin, where the HR value ranged from 0.11 to 0.18. The health risk in the Inner Basin remained the highest for all age groups, especially for infants and adults (HR > 3).

Sensitivity Analyses

Uncertainties in the distribution of surface water fluoride concentration, concentration–response functions and valuation methods affected the health risk estimates. Monte Carlo simulations were adopted to estimate 95% uncertainty intervals from 5000 draws of parameters and concentrations throughout the health risk assessment (Yin et al. 2021a). Crystal Ball software was used for simulation and sensitivity analysis to determine the parameters contributing to the uncertainty of health risk assessment. The higher the value of sensitivity indicates, the greater influence on the uncertainty.

The sensitivity of health risk to uncertain factors are shown in Fig. 8. For dermal absorption, fluoride concentration (C) and exposure time (ETs) contributed the most (over 90%) for the uncertainty, while the exposure time had little influence in all age groups. Among them, the sensitivity of exposure time for residents < 18 years old is two times more than that of adults. Uncertainty in ingestion parameters is in the following order: drinking rate (IR) > fluoride concentration (C) > body weight (BW) > exposure frequency (EF). Therefore, the accuracy of fluoride concentration, exposure time, and drinking rate are key parameters affecting the precision of health risk assessment.

Potential Affected Population

The populations in both the Uniform-type areas and the Scatter-type areas had a low health risk in terms of surface water fluoride. However, endorheic lakes were the main type of water body in the Inner-type areas, and residents living the Inner-type area were more likely to suffer from diseases related to fluoride for the high health risk. Government needs to adopt measures for residents with high health risk in surface water fluoride, such as searching for low fluoride groundwater or helping residents relocate to a place with low health risk in surface water fluoride. For rivers in the TP, the fluoride concentration was less than 1 mg L^{-1}. It is crucial to obtain low fluoride risk source water for Asian countries whose rivers originating in the TP.

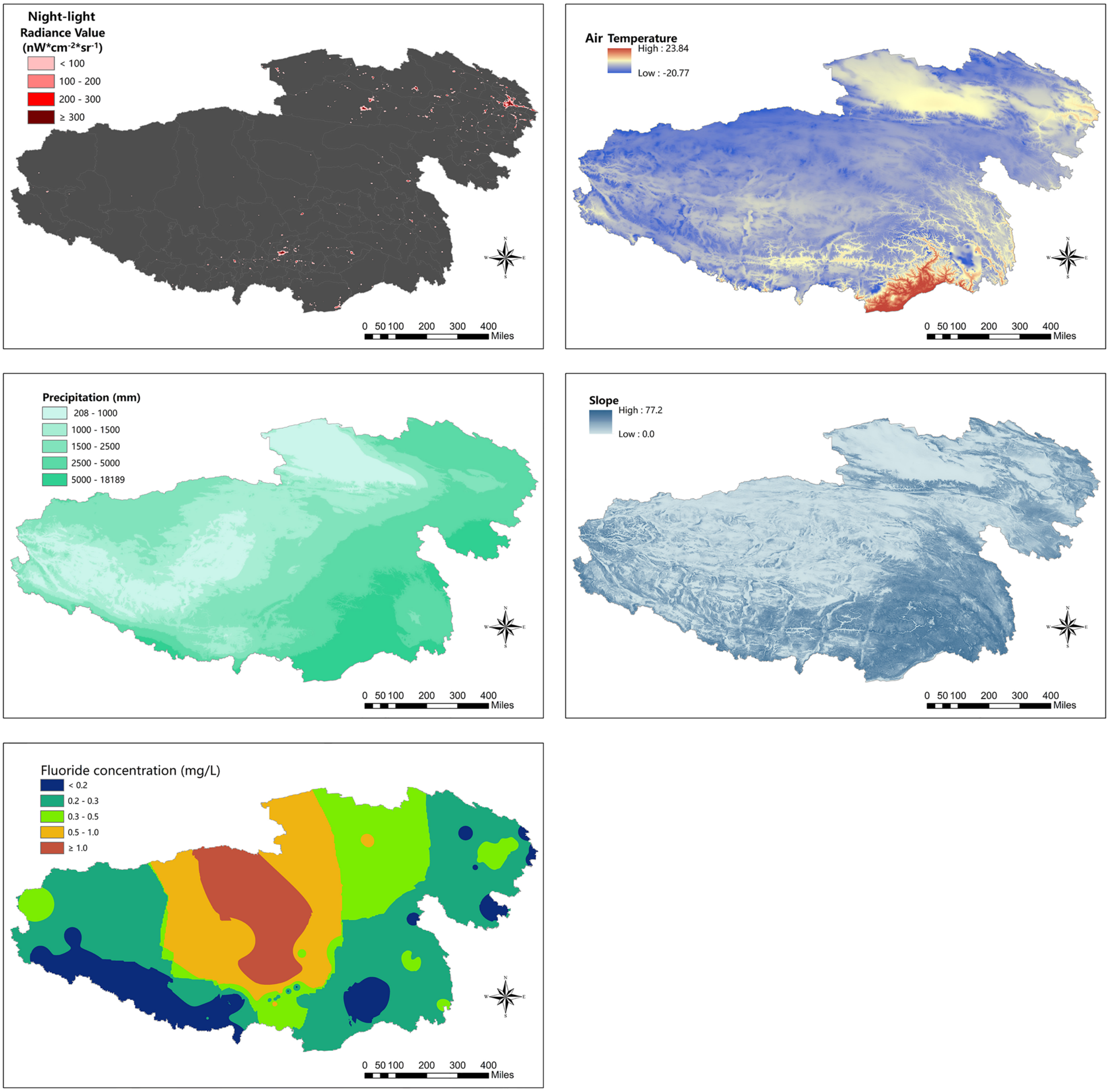

Fig. 6 Distribution of the night light index, air temperature, precipitation, slope, and spatial interpolation of fluoride concentration

Conclusion

The fluoride concentration in surface water in the TP ranged from 0.003 to 6.240 mg L^{-1}. In terms of temporal, fluoride concentrations among dry, flat and rich periods shown insignificant difference, while a tendency for decreasing fluoride concentration by year in the surface water in the TP was obviously present. From spatial perspective, concentration among water basins were in the following order: Inner > 1 mg L^{-1} > Qaidam > Mekong > Salween > Yellow River > Indus > Yangtze > Brahmaputra > Hexi > Ganges; and lakes > rivers.

In general, the fluoride concentration in the surface water in the TP was positively related to anthropogenic activities and was negatively correlated with regional climate factors (precipitation and air temperature). The topography showed an uncertain relationship with fluoride concentration. Consequently, the distribution of the fluoride concentration in the surface water in the TP is compounded by antagonistic and synergistic effects of various factors including anthropogenic activities, regional climate factors and topography, but the

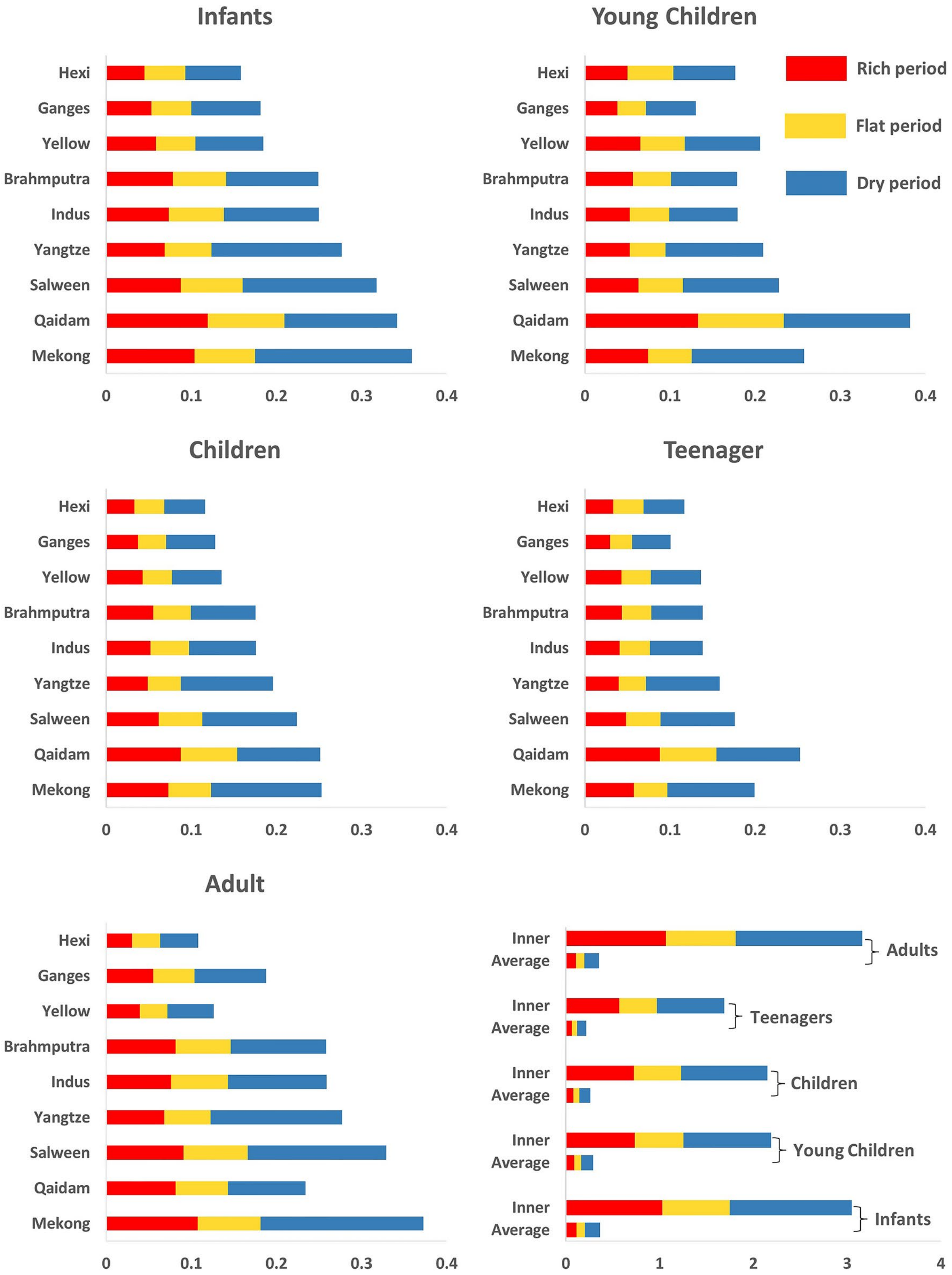

Fig. 7 Health risk by age in different basins

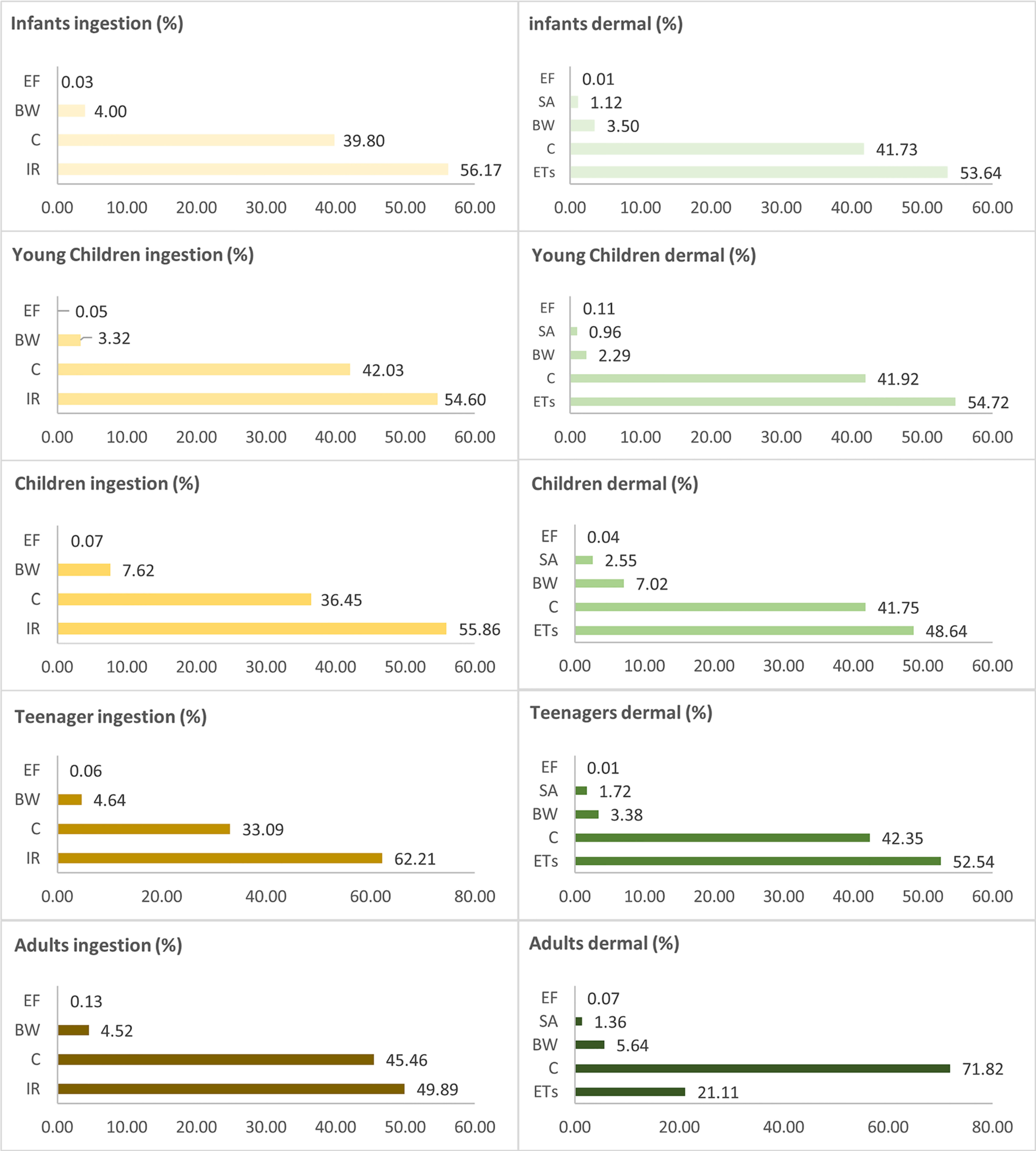

Fig. 8 Sensitivity analysis of age groups for health risk assessment of surface water ingestion and dermal contact

specific mechanism of this compound influence needs to be further studied.

The health risk posed by surface water fluoride ranged 0.07 to 4.61, and ingesting is the main exposure pathway. From age perspective, adults and infants faced more risk. As for water basins, unacceptable risks occurred in the Inner Basin. In addition, Asian countries whose rivers originating in the TP can benefit from the source water with low fluoride health risk.

Nevertheless, our study has some limitations, and further research is needed. Firstly, although the water sampling data in our study cover the main streams and typical lakes of the TP, it cannot represent the region with a high accuracy due to the large number of tributaries. In addition, there are many other sources of human fluoride intake in addition to surface water, such as groundwater and meltwater, which may result in an underestimation of the risk posed by total water fluoride to human health. Therefore, it should be noted that the present study may have underestimated the risk posed by water fluoride to human health in the plateau area.

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Data Availability All the data used for the study appear in the article.

Declarations

Conflict of interest The authors have no relevant financial or non-financial interests to disclose.

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