

Comparative study of physico-chemical parameters of drinking water from some longevity and non-longevity areas of China

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ABSTRACT

There is an obvious regional longevity phenomenon in China and many longevity counties are located in South China. This study was carried out to find the characteristics of elemental contents of drinking water in longevity areas in South China and the differences to non-longevity areas in China. A total of 128 drinking water samples were collected from longevity areas in South China ($n = 40$), non-longevity areas in South China ($n = 74$) and non-longevity areas in North China ($n = 14$) and 46 parameters of water were determined or calculated. The results showed that drinking water in longevity areas of South China had a high ratio of sum concentration of essential micro-elements in sum concentration of micro-elements (SCME) and a low ratio of sum concentration of hazardous micro-elements in SCME. The concentration of total hardness (TH) and strontium in drinking water was 157.82 mg/L and 82.1 $\mu\text{g/L}$, respectively, and they were 14.61 mg/L, 7.45 $\mu\text{g/L}$ and 291.69 mg/L, 748.65 $\mu\text{g/L}$ in the non-longevity areas of South and North China, respectively. The study concluded that drinking water containing 157.82 mg/L TH and 82.1 $\mu\text{g/L}$ strontium in South China may be optimum to human health.

Key words | drinking water, longevity, macro-element, micro-element, South China

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INTRODUCTION

Water is an important component of the human body, providing many essential macro- and micro-elements and playing an important role in human health (Nkono & Asubiojo 1997). Hamilton *et al.* (1973) found a similarity between the abundance of elements present in human tissues and average crystal rocks. Drinking water is also important for the environmental influences of human health (Nkono & Asubiojo 1997).

Increasing attention is being paid to the quality of drinking water worldwide. Water quality can be affected by the natural background and anthropogenic activities. Natural sources of elements are largely the result of chemical weathering of rock or volcanic activity, both of which can have considerable spatial variability (Ritter *et al.* 2002). In some rural areas in China, there is no official tap water and

local residents usually drink water from mountain springs or river water. The quality of drinking water in these areas has a long-term impact on local residents' health.

The chemical and physical properties of elements influence their availability for uptake by biological organisms and their potential to be toxic to organisms, as well as transport mechanisms in natural waters (Ritter *et al.* 2002). Drinking water contains various micro-elements, some of which have been widely implicated in human health and disease (Kumar *et al.* 2016). Elements such as fluorine (F), iron (Fe), manganese (Mn), copper (Cu), cobalt (Co), molybdenum (Mo), zinc (Zn), chromium (Cr), selenium (Se), nickel (Ni), vanadium (V), and silicon (Si) are essential for the growth of human and other living organisms. Even though some of the micro-elements are essential to man, at

elevated levels both essential and non-essential elements can pose a hazardous risk to humans (WHO 1996). Elevated level of F in drinking water can result in problems of increased dental caries and of the development of bone deformities (Zhang *et al.* 2007), and long-term exposure to arsenic in drinking water at elevated levels can result in skin problems, kidney and lung cancers (Smith *et al.* 2002).

There is an obvious regional longevity phenomenon in China (Xin-min 2006). The distribution of centenarians in China is very uneven, and the longevity counties are geographically concentrated in South China. Bama, Donglan, Fengshan, Rugao, Chengmai, Lingao, and San-shui are famous longevity counties in South China. The occurrences of centenarians (OC) (the number of centenarians per 100,000 inhabitants) in these counties are higher than the average level of China and most of the other counties in China. The elemental contents of drinking water in longevity counties in China have been studied by many researchers, but the study areas have always been limited to one longevity county. Until now, the characteristics of elemental contents of drinking water in longevity areas of South China and the difference to non-longevity areas in China have not been clear. Therefore, this study was carried out to investigate the elemental contents of drinking water in the longevity areas of South China and the differences compared to the non-longevity counties in China.

MATERIALS AND METHODS

Natural geographic setting

China is divided into South and North China by Kunlun-Qinling Mountain and the Huaihe River. In South China, the annual precipitation is higher than 800 mm, and the climate belongs to the subtropical monsoon generally. The annual precipitation in North China is lower than 800 mm and the climate belongs to the temperate continental monsoon generally. The study areas are located in South China, except for Huxian, Xinxiang, and Xilinhot counties which are located in North China. More details of the natural geography of those counties are shown in Table S1 (available with the online version of this paper).

Sample collection

A field survey was conducted to collect the drinking water samples from longevity and non-longevity counties of South China. For comparative analysis the drinking water was also collected from the non-longevity counties of North China. First, the percentage of centenarian to county levels was calculated from the national population census (NBS 2010). A total of 128 samples (Table 1) was collected that included 86% of groundwater (spring water and well water) and 14% of surface water (lake water, river water, and reservoir water). Samples collected from longevity areas accounted for 31% and the other 69% was collected from non-longevity areas.

Samples were collected from the households of local inhabitants or from potable water sources (Figure 1). All the samples were collected far away from industrial and mining enterprises and the water sources had not changed for many years. No water pollution had been found according to the local inhabitants and previous literature.

The samples were collected in colorless polythene plastic bottles that had been washed once with distilled water and three times with sample water; this way the chances of contamination in the sample collection would be small. All water samples were stored in clean plastic bottles at 4 °C before further analysis. Details of the sampling method can be found in *Methods for Chemical Analysis of Water and Waste Water* (MEPPRC 2002).

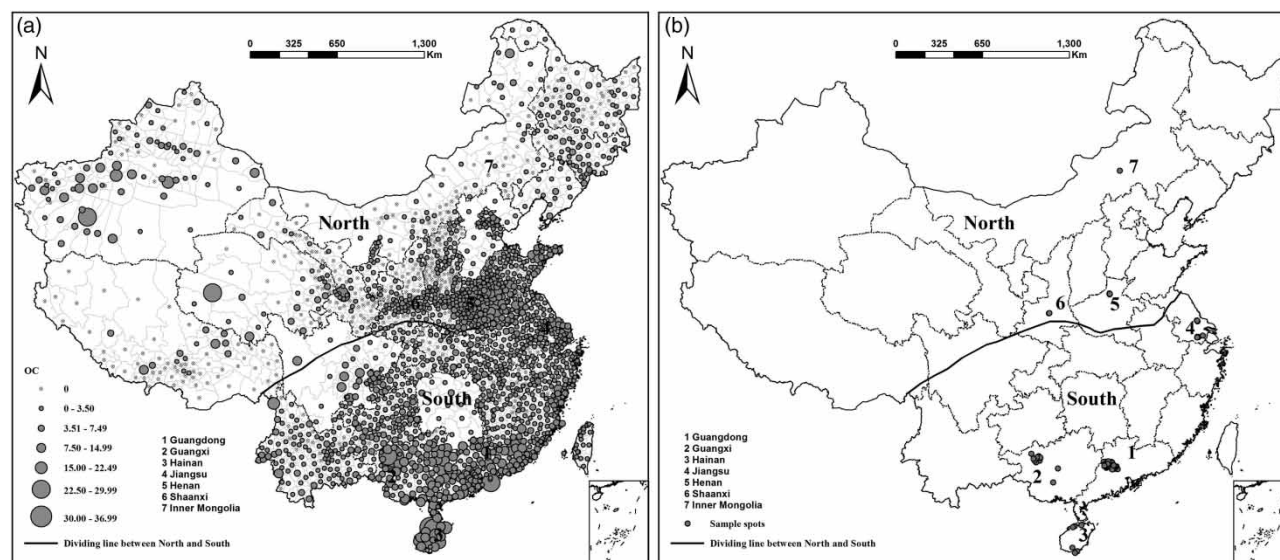
Analytical methods

The concentrations of Ca, Mg, Na, K, SO_4^{2-} , SiO_2 , B, P, Sr were determined by inductively coupled plasma optical emission spectrometry (ICP-OES) (PerkinElmer, Optima 5300 DV) and elements Al, Ba, Be, Cd, Co, Cr, Cu, Li, Mn, Ni, Pb, Tl, V, Cs, Ga, In, Rb, U, Bi, Mo, Zn, and Fe were tested by ICP-mass spectrometry (ICP-MS) (PerkinElmer, Elan DRC-e). The pH, electrical conductivity (EC), and oxidation–reduction potential (Eh) were determined *in situ* using a SevenGo SG2/3 analyzer (Mettler Toledo Company, Shanghai, China). Bicarbonate (HCO_3^-) was determined using an acid–base titration method within 3 days of sampling. Nitrate (NO_3^-) and chlorine (Cl) concentrations in water were determined by ion chromatography (Dionex

Table 1 | Number of samples in the study areas

	Province	County	Lake water	Reservoir water	River water	Spring water	Well water	Total
North	Henan	Xinxiang					6	6
	Inner Mongolia	Xilinhot					1	1
	Shaanxi	Huxian					7	7
South	Guangdong	Guangning				31		31
		Huaiji			1	12	5	18
		Sihui	1			4	5	10
	Guangxi	Sanshui*			1		6	7
		Bama*			1	3		4
		Donglan*	3		1	9		13
		Fengshan*			2	1	1	4
		Heshan					1	1
		Nanning			1			1
		Tianyang						1
	Hainan	Chengmai*					1	1
		Lingao*					4	4
		Sanya			1	1	3	5
Jiangsu	Rugao*		1	1		5	7	
	Suzhou	4				3	7	
Total			4	5	9	62	48	128

*Represents longevity counties.

**Figure 1** | Distribution of longevity counties in China and sample spots.

Inc., Dionex-ICS-900). Total dissolved solids (TDS) were equal to the sum of ions except bicarbonate (HCO_3^-) and half of HCO_3^- . Total hardness (TH) was calculated by the concentration of Ca and Mg.

For determining selenium (Se), 10 mL samples and 5 mL concentrated hydrochloric acid were added into

20 mL glass test tubes and heated in a boiling water bath for 40 minutes. For determining arsenic (As), 10 mL samples, 1 mL concentrated hydrochloric acid, and 1 mL reducing agent (2.5% thiourea + 2.5% ascorbic acid) were added, and allowed to stand for 40 minutes. Both Se and As were determined by hydride generation atomic

fluorescence spectrometry (HG-AFS; Beijing Haiguang Instruments Co. Ltd).

The concentrations of elements were calculated by a five-point calibration curve with linearity of all target elements higher than 0.999. The precision of analyses was tested at regular intervals by running duplicate analyses of selected samples and internal standards. The precision and reproducibility were found to be within the confidence limit of 98%. The detection limit of ICP-MS was $<0.1 \mu\text{g/L}$ and that of ICP-OES and ion chromatography $<0.1 \text{ mg/L}$. The detection limit for As and Se was $<0.01 \mu\text{g/L}$. All the samples were determined in the Geochemistry Laboratory of the Institute of Geographic Sciences and Natural Resources Research, Chinese Academy of Sciences.

Statistical analysis

The Mann–Whitney U nonparametric test was applied as a hypothetical test because the distribution of data did not follow normal distribution or logarithmic distribution. Statistical Package of Social Science version 20 and Excel 2010 were used for the statistical analysis.

RESULTS

Longevity situation of the study areas

The OC (the number of centenarians per 100,000 inhabitants) is an important indicator of the longevity (Magnolfi et al. 2007; Stefanadis 2010; Willcox et al. 2010). However, OC was strongly affected by migration and birth rate (Magnolfi et al. 2007; Liu et al. 2014). To maintain OC, four new indices, namely, LI% (ratio of ultranonenarians to those above 65 years old), CI% (ratio of centenarians within the ultranonenarians), UOI% (ratio of people above 80 years old), UC (number of centenarians per 10,000 over 65 year-old subjects) were applied.

The longevity indices of OC, CI%, and UC in Guangxi province and Hainan province are significantly higher than those of other counties of China (Table 2). Bama and Lingao are typical longevity counties of China as almost all their indices are relatively higher. The differences of longevity indices are apparent and all five longevity indices of

South China are all higher than those of North China. A thought-provoking phenomenon is that OC, CI%, and UC increase consecutively from north-west to south-east in the order of Huaiji, Guangning, Sihui, and Sanshui respectively. The OC, CI% and UC in Sanshui are 12.1-fold, 9-fold, and 14.2-fold, respectively, as compared to northern Huaiji.

Physico-chemical parameters of drinking water

Table 3 shows regular parameters and macro-elements of drinking water in longevity areas and non-longevity areas in South China as well as non-longevity areas in North China.

The pH value of drinking water in longevity areas of South China are 6.62 ~ 8.01 with a median of 7.42, indicating drinking water in longevity areas of South China are slightly alkaline. The TDS of drinking water in longevity areas in South China ranges between 35.43 and 1,014.66 mg/L, varying from fresh water ($\text{TDS} < 1,000 \text{ mg/L}$) to brackish water ($1,000 < \text{TDS} < 2,000 \text{ mg/L}$). The TH of drinking water is 20.14 ~ 594.13 mg/L, varying from soft water ($\text{TH} < 75 \text{ mg/L}$) to high-grade hard water ($\text{TH} > 450 \text{ mg/L}$). The order of relative abundance of major cations of drinking water in longevity areas of South China is $\text{Ca} > \text{Na} > \text{Mg} > \text{K}$ (on a mg/L basis), while that of anions is $\text{HCO}_3^- > \text{SO}_4^{2-} > \text{NO}_3^- > \text{Cl}$. The concentration of micro-elements varies greatly (Table 4). The sum concentration of micro-elements (SCME) is 138.26 ~ 2,269.66 $\mu\text{g/L}$ with a median of 358.28 $\mu\text{g/L}$, and the top three micro-elements in decreasing order according to median value are Fe (108.66 $\mu\text{g/L}$) > Sr (82.1 $\mu\text{g/L}$) > Zn (7.97 $\mu\text{g/L}$). The ranges of the sum concentration of essential micro-elements (SCEE: Fe, Zn, Cu, Mn, Cr, Mo, Co, Ni, V, Se) and sum concentration of hazardous elements (SCHE: As, Cd, Pb) are more than one magnitude with a median of 160.17 $\mu\text{g/L}$ and 1.26 $\mu\text{g/L}$, respectively. The ranges of the ratio of SCEE in SCME ($\text{SCEE}_{\text{SCME}}$) and the ratio of SCHE in SCME ($\text{SCHE}_{\text{SCME}}$) are within two magnitudes with a median of 0.4 and 0.035, respectively.

The pH value of drinking water in non-longevity areas in South China is 5.17 ~ 9.18 with a median of 7.07, indicating drinking water in non-longevity areas in South China is slightly alkaline or weakly acidic. The samples of drinking water in non-longevity areas in South China are all fresh water with TDS ranges between 18.41 and 719.28 mg/L.

Table 2 | Longevity indices of study counties in China

Province	County and city	OC	LI%	CI %	UOI%	UC
Guangdong	Huaiji	0.74	2.27	0.4	1.46	0.92
	Guangning	2.59	1.89	1.24	2.18	2.34
	Sihui	5.16	3.06	2.09	1.86	6.38
	Sanshui	8.99	3.62	3.59	1.68	13.03
Guangxi	Fengshan	28.49	3.41	8.41	1.7	28.71
	Donglan	31.45	4.1	6.33	2	25.95
	Bama	35.61	4.24	9.32	1.65	39.54
	Nanning	1.54	2.13	1.07	1.14	2.27
	Heshan	5.24	2.82	2.09	1.68	5.89
	Tianyang	4.15	2.06	2	1.72	4.1
Jiangsu	Suzhou	1.5	1.99	0.97	1.61	1.93
	Rugao	8.21	2.35	2.08	3.61	4.89
Hainan	Chengmai	31.04	3.68	8.51	2.31	31.32
	Lingao	36.93	4.52	9.5	2.09	42.96
	Sanya	3.21	2.13	3.21	0.75	6.83
Shaanxi	Huxian	0	1.03	0	1.15	0
Henan	Xinxiang	0.88	1.92	0.62	1.44	1.2
Inner Mongolia	Xilinhot	0.58	1.19	0.58	1.2	0.69
South China		3.01	1.80	1.82	1.68	3.28
North China		2.24	1.47	1.77	1.42	2.61
China		2.67	1.66	1.8	1.57	3.0

OC: number of centenarians per 100,000 inhabitants; LI: ratio of ultranonenarians to those above 65 years old; CI: ratio of centenarians within the ultranonenarians; UOI: percentage of people above 80 years old; UC: number of centenarians per 10,000 over 65-year-old subjects.

The TH of drinking water is 0.63 ~ 471.15 mg/L, varying from over two orders of magnitude from soft water to hard water. The order of relative abundance of major cations of drinking water in non-longevity areas in South China is Na > Ca > Mg > K (mg/L), while that of anions is HCO₃⁻ > Cl > SO₄²⁻ > NO₃⁻. The ranges of SCME, SCEE, SCHE, SCHE_{SCME} are no less than two magnitudes with a median of 204.53 µg/L, 75.16 µg/L, 0.96 µg/L, 0.004, respectively. The ranges of SCEE_{SCME} are within one magnitude with a median of 0.35. The top three micro-elements in decreasing order according to median value are Fe (42.04 µg/L) > Al (26.99 µg/L) > Ba (22.71 µg/L).

The pH value of drinking water in non-longevity areas in North China is 7.42 ~ 8.38 with a median of 7.71, indicating drinking water in longevity areas in North China is slightly alkaline. The TDS of drinking water in longevity areas in North China ranges between 172.16 and 1,725.65 mg/L, varying from fresh water to brackish water. The TH of drinking water is 59.2 ~ 755.81 mg/L, varying from over two orders of magnitude from soft water to high-grade hard

water. The order of relative abundance of major cations of drinking water in non-longevity areas in North China is Na > Ca > Mg > K (mg/L), while that of anions is HCO₃⁻ > Cl > SO₄²⁻ > NO₃⁻. The concentration of micro-elements varies greatly. The ranges of SCME, SCEE, SCHE, SCEE_{SCME} are within one magnitude with medians of 1,171.79 µg/L, 158.65 µg/L, 17.12 µg/L, 0.14, respectively. The range of SCHE_{SCME} is within one magnitude with a median of 0.01 and the top three micro-elements in decreasing order according to median value are Sr (748.65 µg/L) > B (90.75 µg/L) > Fe (80.1 µg/L).

Drinking water quality

The quality of drinking water was evaluated based on recommended standards of the World Health Organization (WHO 2011) and National Standards for Drinking Water Quality of the People's Republic of China (MHPRC 2006).

In the longevity areas of South China, TDS, TH, As, Cr, Tl, Zn, and NO₃⁻ are 2.5%, 7.5%, 7.5%, 2.5%, 2.5%, 5%, and

Table 3 | Statistical summary of physico-chemical parameters in drinking waters of China

Elements	Longevity in South n = 40		Non-longevity in South n = 74		Non-longevity in North n = 14		GB 5749-2006 (MHPRC 2006)	WHO (WHO 2011)
	Range	Median	Range	Median	Range	Median		
pH	6.62 ~ 8.01	7.42	5.17 ~ 9.18	7.07	7.42 ~ 8.38	7.71	6.5–8.5	6.5–9.5
EC	37.1 ~ 1,536	329	10.33 ~ 1,146	40.6	286 ~ 2,430	699.50		
Eh	195 ~ 337	231	120 ~ 355	252	193 ~ 290	225		
TDS	35.43 ~ 1,014.66	209.02	18.41 ~ 719.28	47.97	172.16 ~ 1,725.65	441.44	1,000	1,000
TH	20.14 ~ 595.13	157.82	0.63 ~ 471.15	14.61	59.2 ~ 755.81	291.69	450	500
HCO ₃ ⁻	28.3 ~ 737.74	189.67	3.58 ~ 459.97	21.57	151.75 ~ 662.96	337.48		
Ca	6.44 ~ 138.1	50.84	0.17 ~ 125.8	2.63	21.31 ~ 120.5	49.56		
K	0.05 ~ 62.52	1.31	0.13 ~ 17.78	1.45	0.31 ~ 8.35	1.28		
Mg	0.95 ~ 71.89	2.89	0.05 ~ 38.15	1.77	1.46 ~ 128.1	31.02		
Na	0.28 ~ 122.3	4.56	0.35 ~ 76.4	2.95	28.67 ~ 241.3	51.63	200	200
SiO ₂	2.64 ~ 48.17	9.89	2.02 ~ 34.55	17.70	16.35 ~ 28.49	19.26		
Cl	0.84 ~ 148.57	3.26	0.16 ~ 112.91	1.86	<0.1 ~ 401.06	35.73	250	250
SO ₄ ²⁻	0.2 ~ 124.9	7.43	<0.1 ~ 102.9	1.70	5.23 ~ 639.9	35.44	250	500
NO ₃ ⁻	<0.1 ~ 121.93	5.15	<0.1 ~ 90.28	1.56	0.13 ~ 51.59	0.57	44.3	50

Unit: mg/L except EC (μ S/cm) and Eh (mv).

n: number of samples.

TDS: total dissolved solids; TH: total hardness.

7.5%, respectively, higher than the highest limits set by the National Standards for Drinking Water Quality of the People's Republic of China (MHPRC 2006), while their highest values are 1.01-fold, 1.32-fold, 5.63-fold, 1.01-fold, 1.98-fold, 1.95-fold, and 2.75-fold, respectively, as compared with the National Standards for Drinking Water Quality of the People's Republic of China (Tables 3 and 4).

In non-longevity areas of South China, TH, As, Mn, Tl, and NO₃⁻ are 1.35%, 12.16%, 4.05%, 6.76%, and 11.63%, respectively. These parameters in drinking water samples were higher than the highest limits of the standards set by the National Standards for Drinking Water Quality of the People's Republic of China (MHPRC 2006), while their highest values are 1.05-fold, 5.64-fold, 12.13-fold, 1.77-fold, and 3.86-fold respectively, as compared with the National Standards for Drinking Water Quality of the People's Republic of China. The pH in 20.27% of drinking water in non-longevity areas in South China is lower than the lowest limit of 6.5, and the highest and lowest values are 1.08-fold and 0.88-fold, respectively, by comparison with the highest limit (pH = 8.5) and lowest limit (pH = 6.5).

In non-longevity areas of North China, TDS, TH, As, Fe, Na, SO₄²⁻, Cl⁻ and NO₃⁻ are 14.29%, 28.57%, 78.57%, 7.14%, 14.29%, 14.29%, 14.29%, and 11.11% of drinking water samples are higher than the respective limits set by the National Standards for Drinking Water Quality of the People's Republic of China (MHPRC 2006) and their highest values are 1.73-fold, 1.68-fold, 4.06-fold, 1.41-fold, 1.21-fold, 2.56-fold, 1.6-fold, and 1.16-fold, respectively, when compared with the National Standards for Drinking Water Quality of the People's Republic of China.

DISCUSSION

Variations between longevity and non-longevity areas' drinking water quality

The parameters of drinking water from longevity areas and non-longevity areas in South China varied greatly. Mann-Whitney U nonparametric test was applied and the results showed significant difference ($p < 0.05$) in pH (1.05-fold,

Table 4 | Statistical summary of micro-elements in drinking waters of China

Elements	Longevity in South n = 40		Non-longevity in South n = 74		Non-longevity in North n = 14		GB 5749-2006 (MHPRC 2006)	WHO (WHO 2011)
	Range	Median	Range	Median	Range	Median		
Se	<0.01 ~ 7.29	0.12	0.02 ~ 2.28	0.47	<0.01 ~ 9.07	0.25	10	10
As	0.12 ~ 56.33	1.09	0.05 ~ 7.46	0.72	0.92 ~ 40.6	16.95	10	10
Al	<0.1 ~ 109.02	6.04	0.86 ~ 1,127.24	26.99	1.44 ~ 52.17	5.8	200	200
B	<0.1 ~ 209.4	4.9	<0.1 ~ 80.7	<0.1	15.4 ~ 222.2	90.75	500	500
Ba	1.75 ~ 56.98	6.62	0.63 ~ 195.04	22.71	19.99 ~ 359.69	52	700	
Be	<0.1 ~ 0.07	<0.1	<0.1 ~ 1.35	<0.1	<0.1 ~ 0.28	0.01	2	
Cd	<0.1 ~ 1.66	0.02	<0.1 ~ 0.51	0.02	0.01 ~ 0.13	0.04	5	3
Co	0.01 ~ 0.56	0.07	<0.1 ~ 18.24	0.03	0.03 ~ 0.63	0.07		
Cr	<0.1 ~ 50.71	5.93	0.4 ~ 39.22	3.27	3.49 ~ 17.42	9.65	50	50
Cu	<0.1 ~ 4.25	0.19	<0.1 ~ 7.47	<0.1	0.28 ~ 4.34	0.74	1,000	1,000
Li	<0.1 ~ 36.71	0.82	0.05 ~ 9.55	1.21	2.09 ~ 37.22	13.49		
Mn	<0.1 ~ 34.92	0.79	<0.1 ~ 1,213.4	1.01	0.05 ~ 76.66	1.74	100	400
Ni	<0.1 ~ 3.53	0.53	<0.1 ~ 12.9	<0.1	<0.1 ~ 2.76	0.64	20	70
P	<0.1 ~ 227.1	23.3	<0.1 ~ 851.6	<0.1	8.7 ~ 265.8	37.5		
Pb	<0.1 ~ 0.73	0.04	<0.1 ~ 5.03	0.05	<0.1 ~ 0.17	0.01	10	10
Tl	<0.1 ~ 0.2	0	<0.1 ~ 0.18	0.01	<0.1 ~ 0	<0.1	0.1	
V	<0.1 ~ 7.27	0.15	<0.1 ~ 35.91	<0.1	0.45 ~ 43.08	25.66		
Cs	<0.1 ~ 0.85	0.02	0 ~ 0.81	0.06	0 ~ 0.02	0.01		
Ga	<0.1 ~ 4.45	0.26	0.13 ~ 12.36	1.28	0.49 ~ 8.36	1.73		
In	<0.1 ~ 0	<0.1	<0.1 ~ 0.01	<0.1	<0.1 ~ 0.01	<0.1		
Rb	<0.1 ~ 78.88	1.26	0.55 ~ 43.35	3.37	0.09 ~ 1.29	0.39		
U	<0.1 ~ 3.54	0.17	0 ~ 9.84	0.03	0.12 ~ 15.8	0.62		15
Bi	<0.1 ~ 0.01	0	<0.1 ~ 0.07	0	<0.1 ~ 0.01	0		
Sr	20.3 ~ 621.2	82.1	<0.1 ~ 607.3	7.45	177 ~ 2,005	748.65		
Mo	<0.1 ~ 5.1	0.15	<0.1 ~ 5.85	0.07	0.93 ~ 7.6	5.69	70	70
Zn	1.08 ~ 1,951.63	7.97	<0.1 ~ 517.29	13.89	0.33 ~ 197.49	11.99	1,000	3,000
Fe	<0.1 ~ 278.78	108.66	<0.1 ~ 214.85	42.04	33.63 ~ 423.76	80.1	300	3,000
SCME	138.26 ~ 2,269.66	358.28	36.58 ~ 2,691.33	204.53	592.78 ~ 3,205.63	1,171.79		
SCEE	29.73 ~ 2,209.73	160.17	9.13 ~ 13,84.48	75.16	60.51 ~ 550.99	158.65		
SCHE	0.2 ~ 56.35	1.26	0.07 ~ 7.54	0.96	1.13 ~ 40.7	17.12		
SCEE _{SCME}	0.03 ~ 0.98	0.40	0.04 ~ 0.8	0.35	0.07 ~ 0.23	0.14		
SCHE _{SCME}	0.0007 ~ 0.053	0.0035	0.0002 ~ 0.027	0.004	0.0004 ~ 0.058	0.0107		

Unit: µg/L, except SCEE_{SCME} and SCHE_{SCME}.

~: approximate.

n: number of samples.

SCME: sum concentration of micro-elements; SCEE: sum concentration of essential micro-elements (Fe, Zn, Cu, Mn, Cr, Mo, Co, Ni, V, Se); SCHE: sum concentration of hazardous elements (As, Cd, Pb); SCEE_{SCME}: ratio of SCEE in SCME; SCHE_{SCME}: ratio of SCHE in SCME.

median value of longevity areas in South China was 1.05-fold that of non-longevity areas in South China; $p = 0.011$), EC (8.1-fold, $p < 0.001$), major iron HCO_3^- (8.79-fold, $p <$

0.001), Ca (19.36-fold, $p < 0.001$), Mg (1.63-fold, $p < 0.001$), SiO_2 (0.56-fold, $p = 0.014$), SO_4^{2-} (4.38-fold, $p < 0.001$), Cl (1.75-fold, $p = 0.005$), NO_3^- (3.31-fold, $p = 0.028$),

and micro-elements Se (0.25-fold, $p < 0.001$), As (1.58-fold, $p = 0.018$), Al (0.22-fold, $p = 0.002$), B (BD, median value of longevity areas in South China was below detection, $p < 0.001$), Ba (0.29-fold, $p < 0.001$), Co (2.88-fold, $p < 0.001$), Cu (BD, $p < 0.001$), Ni (BD, $p = 0.002$), P (BD, $p < 0.001$), Tl (0.29-fold, $p = 0.004$), V (BD, $p < 0.001$), Cs (0.25-fold, $p < 0.001$), Ga (0.21-fold, $p < 0.001$), Rb (0.37-fold, $p = 0.003$), U (6.55-fold, $p = 0.006$), Sr (11.02-fold, $p < 0.001$), Fe (2.58-fold, $p < 0.001$), SCME (1.75-fold, $p < 0.001$) and SCEE (2.13-fold, $p < 0.001$).

Mann-Whitney U nonparametric test was applied on parameters of drinking water from longevity areas in South China versus non-longevity counties in North China, and the results showed significant difference ($p < 0.05$) in pH (0.96-fold, median value of longevity areas in South China was 0.96-fold that of non-longevity areas in North China, $p < 0.001$), EC (0.47-fold, $p = 0.004$), HCO_3^- (0.56-fold, $p = 0.018$), Mg (0.09-fold, $p = 0.006$), Na (0.09-fold, $p < 0.001$), SiO_2 (0.51-fold, $p = 0.003$), SO_4^{2-} (0.21-fold, $p = 0.009$), NO_3^- (9.03-fold, $p = 0.022$), Se (0.36-fold, $p = 0.003$), As (0.06-fold, $p < 0.001$), B (0.05-fold, $p < 0.001$), Ba (0.13-fold, $p < 0.001$),

Be (0-fold, $p = 0.026$), Cd (0.52-fold, $p = 0.034$), Cu (0.26-fold, $p = 0.009$), Li (0.06-fold, $p = 0.001$), P (0.62-fold, $p = 0.007$), Pb (3.7-fold, $p = 0.012$), Tl (BD, median value of non-longevity areas in North China was below detection, $p = 0.043$), V (0.01-fold, $p < 0.001$), Cs (1.72-fold, $p = 0.014$), Ga (0.15-fold, $p < 0.001$), Rb (3.22-fold, $p = 0.001$), U (0.27-fold, $p < 0.001$), Sr (0.11-fold, $p < 0.001$), Mo (0.03-fold, $p < 0.001$), SCME (0.31-fold, $p < 0.001$), SCHE (0.07-fold, $p < 0.001$), $\text{SCEE}_{\text{SCME}}$ (2.86-fold, $p < 0.001$), and $\text{SCHE}_{\text{SCME}}$ (0.33-fold, $p < 0.001$).

Mann-Whitney U nonparametric test was also applied on parameters of drinking water from non-longevity areas in South China versus non-longevity counties in North China, and the results showed significant difference ($p < 0.05$) in pH (0.92-fold, median value of non-longevity areas in South China was 0.92-fold that of non-longevity areas in North China, $p < 0.001$), EC (0.06-fold, $p < 0.001$), Eh (1.12-fold, $p = 0.007$), HCO_3^- (0.06-fold, $p < 0.001$), Ca (0.05-fold, $p < 0.001$), Mg (0.06-fold, $p < 0.001$), Na (0.06-fold, $p < 0.001$), Cl (0.05-fold, $p = 0.021$), SO_4^{2-} (0.05-fold, $p < 0.001$), As (0.04-fold, $p < 0.001$), B (0-fold, $p < 0.001$), Ba (0.44-fold, $p < 0.001$), Be (0-fold, $p = 0.02$), Cd (0.39-fold,

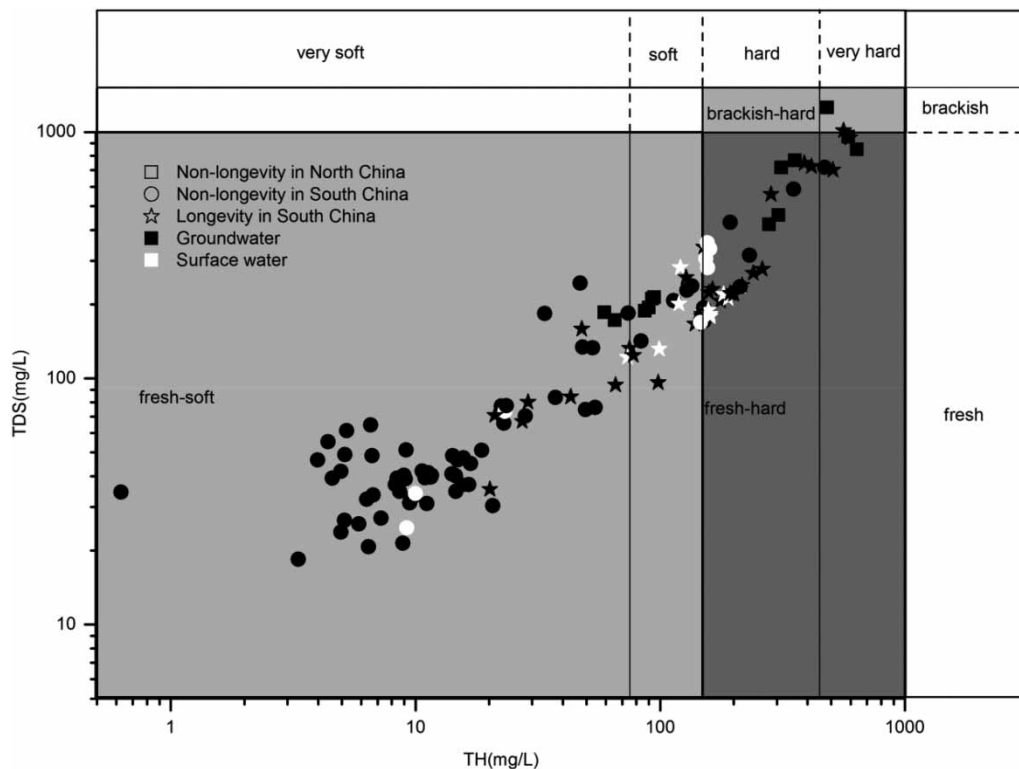


Figure 2 | Drinking water quality in longevity areas and non-longevity areas in South China and non-longevity areas in North China. TDS: total dissolved solids; TH: total hardness.

$p = 0.007$), Co (0.36-fold, $p = 0.001$), Cr (0.34-fold, $p = 0.002$), Cu (0-fold, $p < 0.001$), Li (0.09-fold, $p < 0.001$), Ni (0-fold, $p = 0.006$), P (0-fold, $p < 0.001$), Pb (5.15-fold, $p < 0.001$), Tl (0-fold, $p < 0.001$), V (0-fold, $p < 0.001$), Cs (6.83-fold, $p < 0.001$), In (0-fold, $p = 0.006$), Rb (8.6-fold, $p < 0.001$), U (0.04-fold, $p < 0.001$), Sr (0.01-fold, $p < 0.001$), Mo (0.01-fold, $p < 0.001$), Fe (0.52-fold, $p = 0.009$), SCME (0.17-fold, $p < 0.001$), SCEE (0.47-fold, $p < 0.001$), SCHE (0.06-fold, $p < 0.001$), SCEE_{SCME} (2.5-fold, $p < 0.001$), and SCHE_{SCME} (0.37-fold, $p < 0.001$).

Hydro-geochemical differences

Most of the water samples were fresh water (TDS $< 1,000$ mg/L) (Figure 2) and the others were brackish water ($1,000 < \text{TDS} < 2,000$ mg/L). Fresh water accounted for 97.5%, 100%, and 85.71% of samples from longevity

areas in South China, non-longevity areas in South China, and non-longevity areas in North China, respectively.

Drinking water samples from both longevity areas in South China and non-longevity areas were mainly hard water. The percentage of very soft water (TH < 75 mg/L), soft water ($75 < \text{TH} < 150$ mg/L), hard water ($150 < \text{TH} < 450$ mg/L), and very hard water (TH > 450 mg/L) in longevity areas in South China were 22.5%, 22.5%, 47.5%, 7.5%, and that of non-longevity areas in South China and non-longevity areas in North China were 78.4%, 6.8%, 13.5%, 1.4%, and 14.3%, 28.6%, 28.6%, 28.6%, respectively.

Hydrochemistry type of drinking water samples from longevity areas in South China were Ca-HCO₃⁻ and Ca-Mg-HCO₃⁻ (Figure 3), and in non-longevity areas in South China were Ca-Mg-HCO₃⁻ and Na-K-HCO₃⁻. The major hydrochemistry type of non-longevity areas in North China was Ca-Mg-HCO₃⁻.

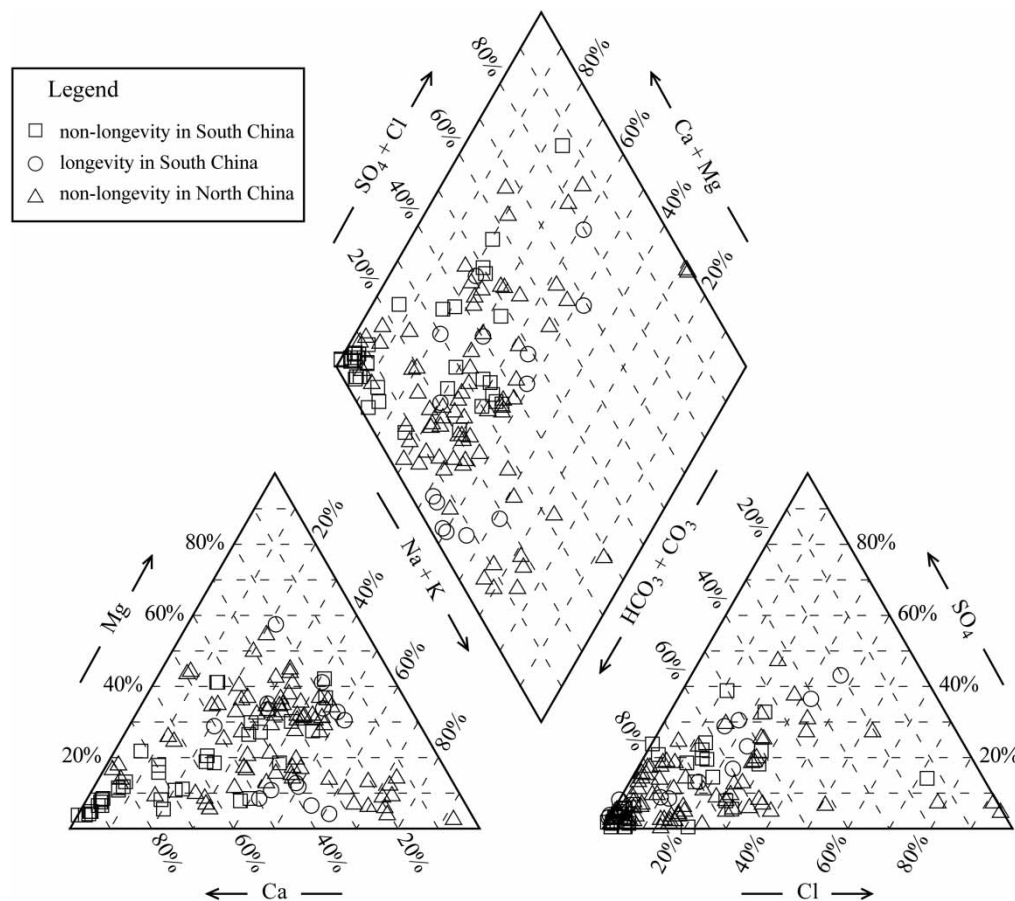


Figure 3 | Piper diagram showing major ion composition of drinking water in longevity areas and non-longevity areas in South China and non-longevity areas in North China.

Relationship between longevity areas and chemical composition of drinking water

The elemental content in drinking water is one of the most important factors for human health and may affect the desirable balance of essential elements in the human body (Keller 1978). Generally, by comparison with non-longevity areas in China, the drinking water in longevity areas of South China has high value $SCEE_{SCME}$ and low value $SCHE_{SCME}$. Compared with non-longevity areas in South China, TH and strontium (Sr) in drinking water in longevity areas in South China were significantly higher. Hard water and very hard water were present in 55% of drinking water in longevity areas in South China, and in 14.9% of drinking water in non-longevity areas in South China (Figure 4). It has been shown that increasing hardness decreases the toxicity of some metals in natural waters to many biological organisms by providing competition for

the metal ions for binding sites in organisms (Ritter *et al.* 2002). A negative relationship between the hardness of drinking water and cardiovascular diseases (CVD) has been found in many published studies (e.g., Monarca *et al.* 2006; Zhang *et al.* 2012). The median concentration of Sr in drinking water in longevity areas in South China was $82.1 \mu\text{g/L}$, which was 11.02-fold higher than that of non-longevity areas in South China ($7.45 \mu\text{g/L}$). In natural water, if the concentration of Sr reaches $200 \mu\text{g/L}$, it would be called mineral water. Strontium is beneficial to the human body in terms of promoting Ca uptake into bone and preventing osteoporosis (Zhang *et al.* 2011; Alegre *et al.* 2012). The elderly are a high-risk group who suffer from CVD and osteoporosis (Hannan *et al.* 2000; Tekin *et al.* 2011). In the longevity areas of South China, the concentration of TH and Sr in drinking water was 157.82 mg/L and $82.1 \mu\text{g/L}$, respectively, which may be the optimum level for human health. Elevated levels of TH

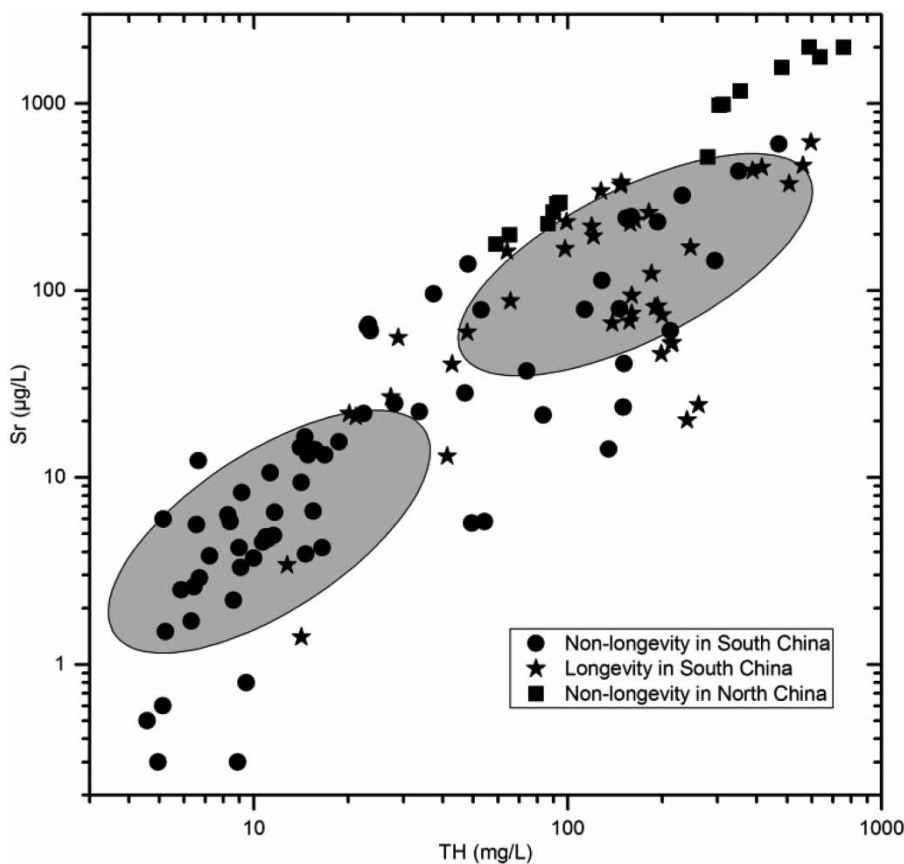


Figure 4 | Sr versus TH of drinking water samples in longevity areas and non-longevity areas in South China and non-longevity areas in North China.

and Sr were found in non-longevity areas in North China, but the SCHE and SCHE_{SCME} in the non-longevity areas of North China were much higher than that of the longevity areas in South China, and SCEE_{SCME} was much lower than that of longevity areas in South China, which may have a potential impact on human health.

The concentrations of As were decreased gradually in Huaiji, Guangning, Sihui, and Sanshui in the order of north-west to south-east, which were in agreement with OC, CI%, and UC, while Cl was the opposite (Table S2, available with the online version of this paper). The concentrations of As and Cl in drinking water in Sanshui were 0.3-fold and 12.35-fold that of Huaiji, respectively.

CONCLUSION

The drinking water of both longevity areas in South China and non-longevity areas in North China was mainly hard fresh water, while that in non-longevity areas in South China was mainly soft fresh water. Most median concentrations of elements in non-longevity areas in South China were lower than those of longevity areas in South China and non-longevity areas in North China. Generally, by comparison with non-longevity areas in China, the drinking water in longevity areas of South China has high value SCEE_{SCME} and low value SCHE_{SCME}. Compared with non-longevity areas in South China, the concentrations of Sr and TH in drinking water in longevity areas of South China were significantly higher, while the same trend was found in non-longevity areas in North China. In the longevity areas of South China, the concentration of TH and Sr was observed at 157.82 mg/L and 82.1 µg/L, respectively, and considered as the optimum level for human health. The elemental concentrations of As were decreased gradually in Huaiji, Guangning, Sihui, and Sanshui in the order of north-west to south-east in agreement with OC, CI% and UC, while Cl was the opposite.

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