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Concentrations of asbestos fibers and metals in drinking water caused by natural crocidolite asbestos in the soil from a rural area

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Abstract Asbestos fibers and metals in drinking water are of significant importance to the field of asbestos toxicology. However, little is known about asbestos fibers and metals in drinking water caused by naturally occurring asbestos. Therefore, concentrations of asbestos fibers and metals in well and surface waters from asbestos and control areas were measured by scanning electron microscopy (SEM), inductively coupled plasma (ICP) optical emission spectrometer, and ICP–mass spectrometry in this study. The results indicated that the mean concentration of asbestos fibers was 42.34 millions of fibers per liter by SEM, which was much higher than the permission exposure level. The main compositions of both asbestos fibers in crocidolite mineral and in drinking water were Na, Mg, Fe, and Si based on energy dispersive X-ray analysis. This revealed that the drinking water has been contaminated by asbestos fibers from crocidolite mineral in soil and rock. Except for Cr, Pb, Zn, and Mn, the mean concentrations of Ni, Na, Mg, K, Fe, Ca, and SiO_2 were much higher in both surface water and well waters from the asbestos area than in well water from the control area. The results of principal component and cluster analyses indicated that the metals in surface and well waters from the asbestos area were significantly influenced by crocidolite mineral in soil and rock. In the asbestos area, the mean concentrations of asbestos fibers and Ni, Na, Mg, K, Fe, Ca, and SiO_2 were higher in surface and well waters, indicating that asbestos fibers and the metals were significantly influenced by crocidolite in soil and rock.

Keywords Crocidolite · Asbestos fiber · Metal · Drinking water · Naturally occurring asbestos

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Introduction

Asbestos has been classified as a human carcinogen by the United States Environmental Protection Agency

and the International Agency for Research on Cancer (IARC 1987; USEPA 1993). Occupational exposure to asbestos was associated with incidence of asbestosis, mesothelioma, lung cancer, and other malignant tumor and pleural diseases (Goodman et al. 1999; Manning et al. 2002; Greillier and Astoul 2008; Mensi et al. 2011). Therefore, many studies have been focused on asbestos fiber contamination in air in asbestos-containing work places such as cement sheet manufacturing industry, automobile brake and clutch manufacturing industry, and thermal insulating boards manufacturing (Blake et al. 2003; Bhagia et al. 2010; Kakooei and Marioryad 2010; Panahi et al. 2011).

Epidemiological investigations revealed that environmental exposure to naturally occurring asbestos, which is not commercially mined and used (Lee et al. 2008), also induced high incidence and mortality rates of mesothelioma, lung cancer, and other asbestos-related diseases (Senyiğit et al. 2000; Metintas et al. 2002; Luo et al. 2005; Perkins et al. 2008; Ates et al. 2010). Therefore, a growing attention has been paid to environmental exposure to asbestos derived from naturally occurring asbestos in recent years (Van Gosen 2007; Favero-Longo et al. 2009; Kuntz et al. 2009). Several studies revealed that abundant asbestos fibers were released into the environment by natural weathering and anthropogenic disturbance of asbestos mineral containing rock and soils (Maresca et al. 1984; Buzio et al. 2000; Anastasiadou and Gidarakos 2007; Hendrickx 2009; Culley et al. 2010). However, few studies were conducted to investigate asbestos fibers in drinking water caused by naturally occurring asbestos from a rural area.

Moreover, asbestos mineral usually contains abundant trace elements. Ni, Co, Cr, and Mn were presented in high concentrations in almost all materials rich in chrysotile asbestos (Schreier et al. 1987). In crocidolite mineral, the concentrations of Ni, Cr, and other elements were also higher (Bowes and Farrow 1997). These metals also significantly affect human health and well-being (Shi et al. 2008; Wei and Yang 2010). In addition, sufficient evidence supported that trace elements play an active role in the induction of asbestos cancer in that asbestos was a passive role as a metal carrier (Dixon et al. 1970; Harrison and Heath 1986). However, little is known about concentrations of asbestos fiber and metals in drinking water derived from naturally occurring asbestos.

Therefore, Dayao, a rural area with abundant outcrop natural crocidolite mineral in soil, was selected as the study area in this study. The main objective of the present study was to measure several metals (such as Ni, Pb, Cu, Mn, Fe, Cr, etc.) in natural crocidolite mineral, concentrations of asbestos fibers, and the selected metals in drinking water (include well water or ground water and surface water). In order to investigate if the metals in drinking water were influenced by asbestos, multivariable analysis were used to identify the sources of metals in drinking water samples from the asbestos and control areas.

Materials and methods

Study area

Dayao is situated in the northwest of Yunnan province, China from 100° 53′ E and 25° 33′ N to 101° 42′ E and 26° 24′ N (Fig. 1). It is characterized by a moderate subtropical monsoon climate, a mean annual temperature of 15.6 °C, and a mean annual rainfall of 796 mm. Rainfall is concentrated in summer and autumn.

Approximately 5 % of the Dayao's surface area contains a thin layer of outcrop crocidolite ore, which is called "blue asbestos" by local residents because of its color. The outcrop crocidolite ore is mainly distributed in ShiYang, Xinjie, and Jinbi covering about 200 km^2 (Fig. 1).

In Dayao, very few villagers can drink running water because of economic backwardness. They mainly drink well water and surface water. The waters may be contaminated by asbestos fibers and other pollutants due to naturally occurring asbestos.

Sampling

Twelve crocidolite mineral samples were collected from different sites in Dayao in 2011. About 500 g of asbestos mineral was collected into clean polyethylene bag for each sample. Then, all the samples were transported to the laboratory.

Fifteen drinking water samples, including 10 well water samples (namely groundwater) and five surface water samples, were collected from different villages for asbestos fiber analysis (Fig. 1). Approximately 800 ml water was collected in a clean polyethylene bottle for each sample. The sample cannot be filtered
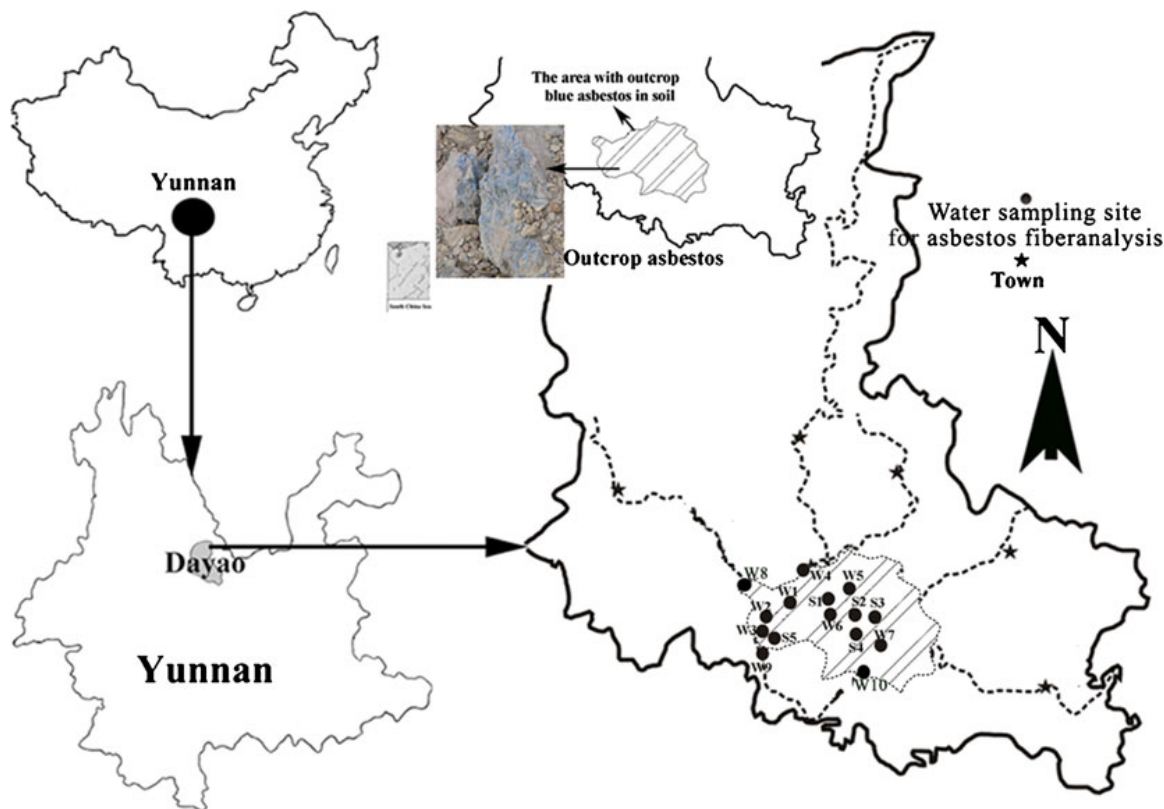
Fig. 1 The map of study area and sampling sites (W1-W10: well water sampling site; S1-S5: surface water sampling site)

within 48 h. Thus, about 1 ml of prefiltered 3 % solution of mercuric chloride was added into each sample to prevent bacterial growth (USEPA 1983).

Moreover, in order to investigate if the metals in drinking water were influenced by natural asbestos, 33 drinking water samples (including 20 well water and 13 surface water) from the area with abundant outcrop asbestos mineral (asbestos area) and 11 drinking water samples from control area were collected. About 100 ml water was collected in a clean polyethylene bottle for each sample.

Analysis method

Asbestos fibers in water samples were determined by scanning electron microscopy (SEM). In the laboratory, the water samples were treated in ultrasonic bath for about 15 min. Then, known aliquots of the samples were filtered through a 0.1-μm pore mixed cellulose ester. A portion of the filter was clipped and treated with gold coating. Then, asbestos fiber on the clipped filter was examined by SEM at a magnification of ×2,000–10,000. The method was conducted according to ISO 14966

(ISO 2002). Main compositions of the asbestos fibers were identified by energy dispersive X-ray analysis (EDXA; USEPA 1983).

The concentrations of Cr, Ni, Pb, Cu, Zn, Mn, Fe, Ca, K, Mg, Na, and Si in water samples were measured by inductively coupled plasma mass spectrometry (ICP-MS) and inductively coupled plasma optical emission spectrometer (ICP-OES).

All the asbestos mineral samples were dried in an oven at 60 °C for 12 h. The dried samples were crushed and sieved via a nylon sieve with a diameter of ≤0.149 mm. Then, approximately 0.1 g of the sieved asbestos sample was digested with 4:8:1 of $HNO_3/HF/HClO_4$ mixture acid. The well-prepared solution was digested at 200 °C. Finally, the molten samples were diluted in a 50-ml flask with deionized water. The concentrations of Cr, Ni, Pb, Cu, Zn, and Mn were analyzed by ICP-MS, while the concentrations of Ca, Fe, K, Mg, and Na were measured by ICP-OES. The concentration of SiO_2 was determined by loss of ignition method. Principal component analysis (PCA) and cluster analysis (CA) of the data were performed by SPSS V18.0 software.

Results

Heavy metals in crocidolite asbestos mineral

The concentrations of the selected metals in crocidolite asbestos mineral are listed in Table 1. The table show that mean concentrations of Cr, Ni, Pb, Cu, Zn, Mn, CaO, Fe_2O_3, K_2O, MgO, Na_2O, and SiO_2 are 34.69, 58.06, 32.19, 20.46, 196.65, 378.62 mg/kg at 1.59, 14.71, 1.01, 7.07, 8.53, and 40.63 %, respectively. It can be seen that the main compositions of crocidolite mineral are Fe_2O_3, MgO, Na_2O, and SiO_2. The background values (CEPA 1990) of the metals in soil in the study area are also listed in the table. Mean values of Ni, Zn, CaO, Fe_2O_3, MgO, and Na_2O in crocidolite mineral were obviously higher than their background values in soil, while the concentrations of the other metals were lower.

Moreover, asbestos fibers and compositions of crocidolite mineral were measured by SEM and EDXA. The photographs of asbestos fibers and image of EDXA are represented in Fig. 2. The figure also indicates that the main compositions of the fiber are Na, Mg, Fe, and Si.

Asbestos fibers in drinking water

The concentrations of asbestos fibers in drinking water (including well and surface waters) in Dayao are listed in Table 2. According to USEPA, the permissible exposure level (PEL) for asbestos fiber in drinking water was seven million fibers per liter (MFL; USEPA 2009). The mean value (42.34 MFL) of asbestos fibers in total drinking water samples exceeded PEL, while the minimum value (0.94 MFL) in well water was lower than

PEL. In Dayao, abundant outcrop crocidolite asbestos mineral was found in soil and rock. Moreover, the results of SEM and EDXA revealed that the main compositions of asbestos fibers in drinking water (Fig. 3) and crocidolite mineral were Fe, Mg, Na, and Si. This may indicate that the drinking water in Dayao has been significantly contaminated by asbestos fibers derived from asbestos mineral in soils and rock.

The table showed that the minimum (15.10 MFL), maximum (399.00 MFL), and mean (109.82 MFL) concentrations of asbestos fibers were all higher in surface water than in well water from asbestos area (Table 2). This reflected that contamination level of asbestos fiber was much higher in surface water than in well water. It may be attributed that the surface water was strongly influenced by asbestos fibers from natural weathering of outcrop crocidolite mineral including wet deposition, dry deposition, and water corrosion of outcrop crocidolite mineral. Moreover, abundant asbestos fibers were released into surface water because of significant disturbance of crocidolite mineral in soil and rock by human activities. The human actives mainly included agricultural activities, urban development, road and building construction, and deforestation. The processes of these activities can directly release abundant asbestos fibers into surface water. Indirectly, asbestos mineral become fine and friable during these activities. Thus, asbestos mineral become more easily weathered and corroded by natural factors. As a result, more asbestos fibers were emitted into surface water. Therefore, the concentrations of asbestos fibers were higher in surface water samples than in well water samples.

Table 1 Concentrations of selected metals in crocidolite asbestos mineral

Element	Minimum	Maximum	Mean	Background value in soil
Cr mg/kg	20.47	65.89	34.69	65.2
Ni mg/kg	23.11	113.49	58.06	42.5
Pb mg/kg	9.7	34.75	32.19	40.6
Cu mg/kg	6.64	51.88	20.46	46.3
Zn mg/kg	45.53	322.26	196.65	89.7
Mn mg/kg	134.13	853.82	378.62	626
CaO %	0.4	4.2	1.59	0.16
Fe_2O_3%	5.9	21.17	14.71	5.22
K_2O %	0.28	2.42	1.01	1.61
MgO %	1.98	15.2	7.07	0.61
Na_2O %	1.26	26.9	8.53	0.16
SiO_2%	18.86	57.46	40.63	

Fig. 2 The photographs of asbestos fibers of crocidolite mineral under SEM and image of EDXA
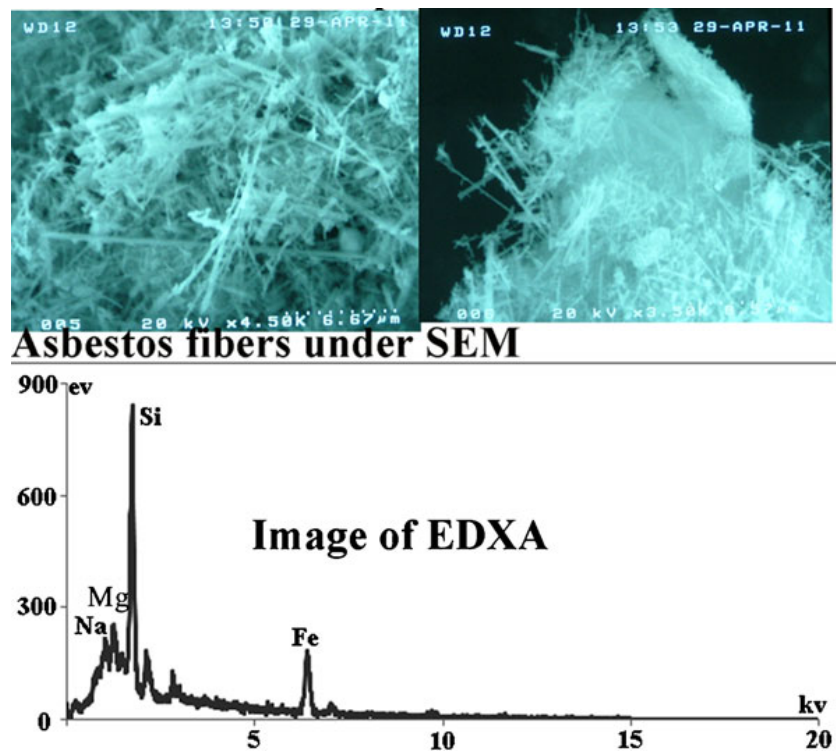
However, well water samples were also significantly contaminated by asbestos fibers. The mean concentration of asbestos fibers in well water samples was higher than the PEL. In Dayao, the wells were all in the open air. Thus, the well water may be also contaminated by asbestos fibers from wet and dry depositions of particle matter in air. In addition, crocidolite mineral layer was usually contained in subsurface soil. The subsurface crocidolite mineral was an outcrop in the shaft wall. Therefore, water corrosion of crocidolite asbestos in the shaft wall may also release abundant asbestos fibers into well waters.

In this study, the mean concentrations of asbestos fibers in surface water was much higher than that in Aliakmonas river caused by deposit tailing of asbestos mine, and in Silver lake and Staten Island lake caused by serpentinite bedrock (Table 2). The mean concentration of asbestos fibers in well water was also higher than in Staten Island Lake. This confirmed that the drinking water in Dayao has been significantly contaminated by asbestos fibers derived from natural crocidolite mineral.
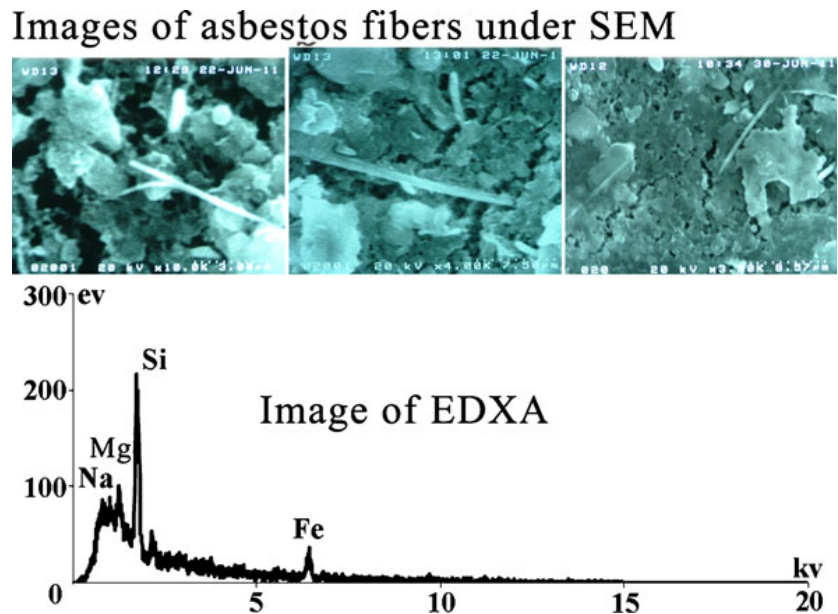
Metals in drinking waters

The concentrations of the selected metals in drinking water in asbestos and control areas from Dayao were represented in Table 3. The highest concentrations of Cr, Ni, Cu, Mn, Fe, Ca, K, Mg, and SiO_2 were found in surface water in asbestos area, while the lowest concentrations of Ni, Cu, Mn, Fe, Ca, K, Mg, and SiO_2 were found in well water in control area. Except for Cr, Pb, Zn, and Mn, the mean concentrations of the other metals were much higher in surface water and well water from asbestos area than in well water from control area. In Dayao, the concentrations of Ni, Zn, CaO, Fe_2O_3, K_2O, MgO, and

Table 2 Concentration of asbestos fibers (millions of fibers per liter (MFL) by SEM) in drinking water

Data	Min. (MFL)	Max. (MFL)	Mean (MFL)	Reference
Well water	0.94	50.20	8.60	This study
Surface water	15.10	399.00	109.82	This study
Total			42.34	This study
Silver lake	16	86	47	Maresca et al. (1984)
Staten Island lake			1.0	Maresca et al. (1984)
Aliakmonas river	5.1	176.2	42.2	Emmanouil et al. (2009)

Fig. 3 The photographs of asbestos fibers in drinking water samples and image of EDXA

Na$_2$O were much higher in crocidolite mineral than their background values in soil. These features may reveal that drinking water in the asbestos area was significantly influenced by the metals derived from outcrop asbestos mineral.

Principal components analysis

PCA was employed to identify possible sources of selected metals in dinking waters from different areas. The principal component loadings, extracted by using varimax normalized rotations on the dataset, are listed in Table 4. Accordingly, three principal components (PCs) were extracted as groups of various metals, with greater eigenvalue against the cumulative total variance of more than 85 % for surface water in asbestos area. PC 1 showed maximum loadings for Ni, Zn, Fe, Ca, Mg, and Na with a significant contribution of K accounting for 44.94 % of variance. These metals may be mainly derived from natural sources. PC 2 was dominated by Cu and K, implying that the metals were mainly from anthropogenic sources. However, PC 3

Table 3 Concentrations of selected metals in drinking water from asbestos area and control area

Metal	Surface water in asbestos area		Well water in asbestos area		Well water in control area	
	Range	Mean	Range	Mean	Range	Mean
Cr µg/L	1.68–8.94	3.97	1.51–7.76	3.48	2.25–6.39	3.75
Ni µg/L	0.51–5.78	3.78	0.82–9.31	3.59	0.31–1.91	1.01
Pb µg/L	0.00–0.11	0.04	0.00–0.07	0.03	0.01–0.09	0.04
Cu µg/L	0.09–1.73	0.81	0.05–2.31	0.46	0.05–1.04	0.38
Zn µg/L	0.44–2.49	1.42	0.44–8.15	3.09	0.27–8.22	3.46
Mn µg/L	0.52–6.29	3.43	0.07–7.12	1.71	0.30–4.59	1.62
Fe mg/L	0.07–0.84	0.50	0.08–1.23	0.45	0.04–0.23	0.11
Ca mg/L	23.85–285.60	167.91	21.77–423.80	142.76	13.17–68.88	32.99
K mg/L	1.20–10.54	5.29	0.38–21.96	3.46	0.74–9.62	2.09
Mg mg/L	19.40–74.09	43.41	10.97–67.03	31.06	2.67–29.10	14.50
Na mg/L	12.94–39.58	24.42	7.80–89.32	28.88	4.51–35.42	13.69
SiO$_2$ mg/L	5.02–22.66	16.18	2.92–37.86	13.23	2.62–12.45	6.81

Table 4 Principal component loadings for selected metals in the drinking water of three groups

Metal	Components of surface water in asbestos area			Components of well water in asbestos area			Components of well water in control area			
	1	2	3	1	2	3	1	2	3	4
Cr	0.240	−0.021	0.874	0.659	−0.068	−0.130	0.219	−0.032	0.870	0.069
Ni	0.961	0.059	−0.003	0.960	0.046	0.001	0.796	0.215	0.140	0.483
Pb	0.219	−0.465	0.766	0.041	0.935	0.055	0.276	0.911	0.034	0.103
Cu	0.085	0.900	0.161	0.660	−0.148	0.451	0.562	0.562	0.030	0.506
Zn	0.930	−0.147	−0.029	−0.068	0.913	0.020	0.502	−0.568	−0.418	0.450
Mn	0.058	0.084	0.645	−0.069	−0.005	0.755	−0.088	0.917	−0.100	−0.060
Fe	0.910	0.312	0.238	0.948	−0.106	−0.016	0.941	0.136	0.243	−0.049
Ca	0.910	0.308	0.239	0.970	−0.077	−0.067	0.932	0.098	0.265	−0.085
K	0.391	0.884	−0.101	0.433	−0.013	0.462	−0.056	−0.016	0.345	0.895
Mg	0.950	−0.020	0.301	0.917	−0.042	0.004	0.395	−0.282	0.703	0.271
Na	0.881	0.271	0.267	0.914	0.105	0.008	0.756	−0.306	0.434	0.147
SiO_2	0.025	−0.739	0.515	0.409	−0.387	−0.656	−0.282	−0.424	−0.744	−0.301
Total	5.393	2.653	2.345	5.674	1.917	1.443	3.899	2.739	2.448	1.708
% of variance	44.944	22.111	19.539	47.282	15.973	12.025	32.492	22.827	20.401	14.235
Cumulative %	44.944	67.055	86.594	47.282	63.255	75.280	32.492	55.319	75.720	89.955

was dominated by Cr, Pb, and Mn, with high loadings of Na, Mg, and SiO_2, indicating that the metals may be mainly derived from soil.

In the case of well water in asbestos area, PC 1 was dominated by Ni, Fe, Ca, Mg, and Na, with a significant contribution of Cr, Cu, K, and SiO_2, accounting for 47.28 % of total variance. These metals could be attributed to natural sources. Pb and Zn at a total variance of 15.97 % were contributed to PC 2, which could be attributed to anthropogenic sources. PC 3 incorporated a variable 12.03 % of total variance in favor of Mn, Cu, K, and SiO_2.

In comparison, for well water in the control area, four PCs were extracted by PCA with a cumulative variance of more than 89 % (Table 4). PC 1 showed elevated loadings of Ni, Fe, Ca, Na, with strong contribution of Pb, Cu, Zn, and Mg. This could be attributed to natural sources. PC 2 was dominated by Pb, Mn, Cu, and Zn. PC 3 explained about 20.40 % of the total variance, implying that the metals may be derived from anthropogenic sources. It was dominated by Cr, Mg, and SiO_2, with a significant contribution of Zn, Ca, K, and Na. PC 4 was dominated by K, Ni, Cu, and Zn, accounting for 14.24 % of the total variance.

The loadings of Na, Mg, K, Ca, Fe, Zn, and Ni of PC 1 were usually much higher in cases of well and surface waters in asbestos area than in the case of well water in the control area. These findings reflected that the metals, particularly Na, Ni, Mg, K, and Zn, have different main sources in well and surface waters from asbestos area compared with well water from control area. In asbestos area, the outcrop crocodolite contained higher concentrations of Na_2O, MgO, Fe_2O_3, CaO, Ni, and Zn. The concentrations of the metals were also much higher in well and surface waters from the asbestos area than in well water from the control area. These features indicated that the concentrations of Na, Ni, Mg, K, and Zn in well and surface waters from asbestos area may be significantly influenced by outcrop crocidolite mineral in soil and rock.

Cluster analysis

CA was applied to standardize the bulk concentrations of metals using Ward's method by square Euclidian distances. The results are shown as a dendrogram in Fig. 4. In case of surface water from the asbestos area, three major clusters of the selected metals were identified: a significant cluster among Fe-Ca-Ni-Zn-Mg-Na, another cluster including Cu-K, and the third cluster comprising Cr-Pb-Mn-SiO_2. The clustering of

Fig. 4 Cluster analysis of selected metals in the drinking water of three groups
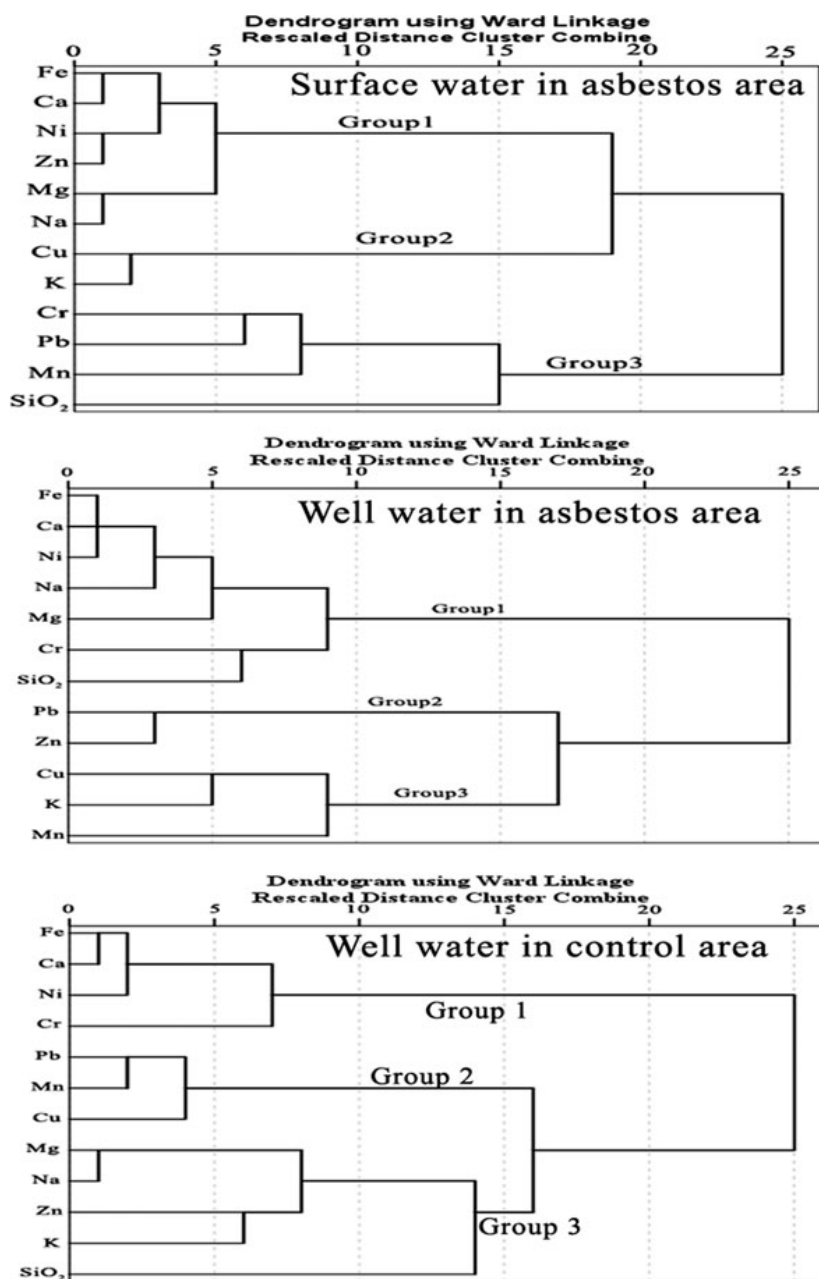
selected metals in well water from asbestos area was slightly different from that of the previous case. Three major clusters were observed among: Fe-Ca-Ni-Na-Mg-Cr-SiO$_2$, Pb-Zn, and Cu-K-Mn. It can be found that Fe, Ca, Ni, Zn, Mg, and Na are all included in the first clusters in both well water and surface water in asbestos area. This confirmed the results of PCA that the metals in both well and surface waters in asbestos area were significantly influenced by crocidolite mineral in soil and rock. However, the first cluster in the well water also contained Cr and SiO$_2$, indicating that

the metals may be less significantly influenced by crocidolite mineral in well water than in surface water from the asbestos area.

In well water from the control area, three major clusters were observed. A very strong cluster was found for Fe-Ca-Ni-Cr. The other two significant clusters were Pb-Mn-Cu and Ng-Mg-Zn-K-SiO$_2$. The results revealed that the clustering of the selected metals in well water from the control area was significantly different from that for both well and surface waters from the asbestos area. Na, Mg, Zn, K, and

SiO_2 may be mainly derived from soil, while the other metal may be originated from anthropogenic sources such as agricultural actives.

Conclusions

According to SEM, the well and surface waters in the asbestos area were significantly contaminated by asbestos fibers. The results of EDXA indicated that asbestos fibers were mainly derived from crocidolite mineral in soil and rock. Moreover, the contamination level of asbestos fibers was much higher in surface water than in well water in the asbestos area.

The concentrations of Ni, Zn, Fe_2O_3, CaO, MgO, and Na_2O were higher in crocidolite mineral than background values in soil. The concentrations of Ni, Fe, Ca, Mg, and Na were also much higher in surface and well waters from the asbestos area than in well water from the control area. The results of PCA and CA indicated that the metals in surface and well waters from the asbestos area were significantly influenced by crocidolite mineral in soil and rock. Moreover, the selected metals may be more significantly influenced by crocidolite mineral in surface water than in well water in the asbestos area. Therefore, more attention should be paid to investigate the contamination of asbestos fibers combined with metals in drinking water in future.

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