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Selenium in soil and endemic diseases in China

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Abstract

Selenium is an essential element for humans, animals and some species of microorganisms. The biological function of selenium shows dual characteristics. The selenium content range between toxic and deficient concentration is very narrow. The present paper discusses the geographical distribution of two forms (total and water-soluble) of selenium in topsoil (plough layer for cultivated soils, eluvial horizon for natural soils) and evaluates its relationship with some human health problems in China. Topsoil samples, 354 in total, including 156 natural and 198 cultivated soils of 21 main soil types were collected. The total Se concentration in soil samples was determined with DAN (di-aminonaphthalene)-fluorescence spectrophotometer method. Soil water-soluble Se concentration was determined with the same method after extraction with water (water/soil = 5:1). The results showed that the geometric and arithmetic means of total Se concentration in soil, for all samples, were 0.173 mg/kg and 0.239 mg/kg, respectively, with the lowest value being 0.022 mg/kg and the highest being 3.806 mg/kg. For the cultivated soil, the geometric mean of total Se was 0.188 mg/kg, its arithmetic mean was 0.269 mg/kg and higher than those in the natural soil, 0.154 mg/kg and 0.206 mg/kg, respectively. The geometric and arithmetic means of water-soluble Se in soil for all the samples were 4.0 and 6.4 μg/kg, the lowest 0.6 μg/kg and the highest value being 109.4 μg/kg. For the cultivated soils, the average concentration of water-soluble Se was 4.3 μg/kg, similar to that of natural soil, they are and 4.4 μg/kg by geometric mean. Two sequences of the soil types, arranged separately in the concentration of total Se and water-soluble Se, are different and this demonstrates that the proportions of the two forms of selenium existing in various soils are different. The percentages of water-soluble Se to total Se in different types of soils varied from 1.07 to 6.69%. However, generally the laterite and other subtropic soil still have relatively high absolute water-soluble Se contents because of their higher total Se contents. A very significant correlation between total Se and water-soluble Se has been found in cultivated soil with a correlation coefficient of 0.58 ($P < 0.01$). The relationships between soil Se and human endemic diseases Keshan disease, Kashin–Back diseases and selenosis have been discussed. The reference criteria for evaluating Se deficiency and Se excess in soil were suggested.

Keywords: Selenium; Soil; Water-soluble Se; Endemic diseases; Keshan Disease; Endemic cardiomyopathy; Kashin–Beck Disease; Endemic osteoarthropathy; Health

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1. Introduction

Selenium (Se) is an essential trace element for humans, animals and some species of microorganism (Pinsent, 1954; Underwood, 1977; WHO 1987; Oldfield, 1987; Stadtman, 1987). Selenium is an integral part of the enzyme glutathione peroxidase (GSH-Px) in man and animals (Schwarz and Foltz, 1957; Rotruck et al., 1973). This enzyme can protect the organism against oxidative damage by reducing lipoperoxides and hydrogen peroxide. Our previous studies showed that selenium also exists in the glutathione peroxidase in higher plant species such as wheat, maize, grape, soybean and garlic (Xue et al., 1993a,b; Hou et al., 1994). It plays a role of antioxidation through the enzymatic mechanism of glutathione peroxidase, which is similar to that in human and animal organisms. Selenium can also stimulate the seedling growth of the above mentioned plant species, which has never been reported before (Rosenfeld and Beath, 1964; Xue et al., 1993a,b; Hou et al., 1994). Selenium is also incorporated into other enzymes or proteins, such as phospholipid hydroperoxide glutathione peroxidase (PHG-px). Selenoprotein P (Se–p), and type I iodothyronine 5′-deiodinase (5′-ID-I), which all have important human health functions. Several microbiological Se containing enzymes have been also identified, e.g. glycine reductase, formate dehydrogenase, hydrogenase, nicotinic acid hydroxylase, xanthine dehydrogenase and thiolase (Pinsent, 1954; Stadtman, 1987; Tappel, 1987).

The biological function of selenium shows a dual character because the selenium content range between toxic and deficient concentration in animals is rather narrow. Selenium is unevenly distributed on the surface of the earth and consequently the selenium concentration in different geo-ecosystems varies widely, forming seleniferous and Se-deficient geo-ecosystems. This uneven distribution is likely to affect health of both humans and animals through the food chain. Generally the selenium in soil is the primary source of human food Se. A study on selenium content in soils and its existing forms can improve our understanding and evaluating the selenium cycling, flux and balance in geo-ecosystems and their influence on health. The present study discussed the geographical distribution and correlation of total and water-soluble selenium in topsoil, and its effect on human health in China. It would be also of great importance to evaluate the association between the selenium status in the environment and Keshan disease (KSD) and Kashin–Beck disease (KBD), to propose appropriate measures, through the improvement of the Se in soil, to control the endemic diseases, which are related to the abnormal selenium content in soils. The present study on soil selenium has made use of large scale geographically and systematization, compared with other similar studies conducted in China (Cheng et al., 1980; Tan et al., 1987a,b; Cheng et al., 1984).

2. Materials and methods

In total 354 topsoil samples, including 156 natural soils and 198 cultivated soils, were collected from 21 main soil types in the whole country. Soil samples free of plant roots were air-dried, passed through a 100-mesh sieve for total Se (T-Se) determination and through a 20-mesh sieve for water-soluble Se (WS-Se) determination separately and then stored at room temperature. The water-soluble selenium was extracted with distilled deionized water at a ratio of water to soil of 5:1 (W/W) and shook for 30 min on an oscillator. The selenium in samples was all analyzed by DAN (2,3-diaminonaphthalene)-fluorescence spectrophotometer (Hou et al., 1981). The detection limit is 0.002 μg/ml, the recovery rate is 93.5–102.6% with an average of 98.9% and the coefficient of variation was 2.74%. With regard to the statistical methods in present paper, SPSS/PC software was employed for calculating the mean, standard deviation, minimum value, maximum value and the product–moment (Pearson) correlation coefficient of total soil and water-soluble Se concentrations.

3. Results and discussion

The selenium concentrations in soil samples,

Table 1
The total and water-soluble Se concentration in topsoils in China

Type	*N*	X	S.D.	G	S_{lg}	Min	Max
Cultivated							
T-Se (mg/kg)	198	0.269	0.381	0.188	0.3017	0.038	3.801
WS-Se (μg/kg)	171	6.0	7.0	4.3	0.6	0.6	69
Natural							
T-Se (mg/kg)	157	0.206	0.227	0.154	0.3125	0.022	1.697
WS-Se (μg/kg)	93	7.0	15.0	4.4	0.3824	0.9	109
Total							
T-Se (mg/kg)	354	0.239	0.320	0.173	0.3022	0.022	3.806
WS-Se (μg/kg)	264	6.4	9.2	4.0	0.3402	0.6	109.4

Note: T-Se: total Se; WS-Se: water soluble Se; *N*: number of samples; X: arithmetic mean; G: geometric mean; S.D.: arithmetic standard deviation; S_{lg}: geometric standard deviation.

both total and water-soluble, are of a logarithmic normal distribution. Therefore, the geometric mean was used for expressing the average concentration of Se content in soils.

In the present study, both the geometric and arithmetic means of Se concentration for different soil types are listed in Tables 1 and 2. It is shown that the arithmetic means of Se concentration in various soils are always higher than the corresponding geometric means because of the

Table 2
The Se concentration in different soil types in China

Type	*N*	T-Se (mg/kg)		WS-Se (μg/kg)		WS/T-Se%	
		A	G	A	G	A	G
1 Dryland cultivated soil	154	0.287 ± 0.421	0.190 ± 0.3110	6.5 ± 7.8	4.7 ± 0.3164	2.24	2.47
2 Paddy soil	41	0.201 ± 0.089	0.181 ± 0.3323	3.9 ± 2.4	3.1 ± 0.2664	1.94	1.71
3 Laterite	2	0.832	0.946 ± 0.1679	11.0	15.6 ± 0.0156	1.39	1.65
4 Red Soil	15	0.345 ± 0.189	0.315 ± 0.2436	4.2 ± 1.6	3.4 ± 0.2508	1.22	1.08
5 Yellow soil	12	0.549 ± 0.532	0.397 ± 0.3408	7.1 ± 3.1	6.3 ± 0.2621	1.29	1.56
6 Yellow brown soil	13	0.101 ± 0.050	0.089 ± 0.2275	1.8 ± 1.3	1.6 ± 0.1218	1.79	1.80
7 Brown earth	10	0.133 ± 0.099	0.117 ± 0.1970	1.6 ± 0.6	1.6 ± 0.1218	1.33	1.37
8 Drab soil	5	0.098 ± 0.032	0.098 ± 0.1354	1.9	1.9	1.95	1.95
9 Dark brown soil	2	0.096 ± 0.025	0.094 ± 0.1205	2.0	1.9 ± 0.2356	2.09	2.04
10 Grey forest soil	4	0.195 ± 0.054	0.188 ± 0.1364	3.7 ± 1.4	3.5 ± 0.1523	1.90	1.86
11 Red drab soil	9	0.111 ± 0.056	0.103 ± 0.2207	1.8 ± 0.5	1.1 ± 0.1357	1.62	1.07
12 Black soil	2	0.236	0.236	3.2	3.0	1.36	1.28
13 Loessial soil	8	0.103 ± 0.069	0.099 ± 0.1332				
14 Chernozem	4	0.253 ± 0.145	0.223 ± 0.2508	9.8 ± 0.5	8.9 ± 0.0207	3.52	3.99
15 Chestnut soil	12	0.167 ± 0.080	0.147 ± 0.2340	5.0 ± 3.2	4.5 ± 0.2831	2.99	3.06
16 Calcic brown soil	4	0.154 ± 0.072	0.150 ± 0.1919	7.2 ± 3.6	4.0 ± 0.2843	4.50	2.67
17 Meadow soil	20	0.145 ± 0.069	0.126 ± 0.2561	5.0 ± 2.4	4.5 ± 0.2750	3.71	3.57
18 Desert soil	9	0.186 ± 0.091	0.166 ± 0.2331	18 ± 36.8	6.9 ± 0.5513	9.88	4.16
19 Purple soil	7	0.086 ± 0.048	0.076 ± 0.2109	2.2	2.0 ± 0.3107	2.56	2.63
20 Solonchak	10	0.198 ± 0.122	0.169 ± 0.2186	13.1 ± 7.1	11.3 ± 0.2603	6.63	6.69
21 Swamp soil	6	0.122 ± 0.032	0.119 ± 0.1083	2.4	2.4	1.96	2.02

Note: A = arithmetic; G = geometric.

positively skewed distribution of the results, but the two groups of the means have a parallel trend for the soil types.

3.1. Total selenium (T-Se)

The geometric and arithmetic means of total Se concentration for all topsoil samples were 0.173 and 0.239 mg/kg, respectively, the lowest value being 0.022 mg/kg and the highest value being 3.806 mg/kg. For the cultivated soil, the geometric and arithmetic means of total Se concentration were 0.188 and 0.265 mg/kg, higher than those of the natural soils, 0.154 and 0.206 mg/kg, respectively (Table 1). The results show that the total selenium concentrations in soil vary apparently in different soils, depending on the soil types. The Se concentrations in 21 main types of soils studied are the following order (by geometric mean, mg/kg; Table 2):

Laterite (0.946) > yellow soil (0.397) > red soil (0.315) > > black soil (0.236) > chernozem (0.223) > dryland cultivated soil (0.190) > grey forest soil (0.188) > paddy soil (0.181) > solonchak (0.169) > desert soil (0.166) > calcic brown soil (0.150) > > chestnut soil (0.147) > meadow soil (0.126) > swamp soil (0.119) > brown earth (0.117) > red drab soil (0.103) > loessial soil (0.099) > drab soil (0.098) > dark brown soil (0.094) > yellow brown soil (0.089) > purple soil (0.076).

The soils developed under tropic and subtropic conditions, such as laterite, yellow soil and red soil, have higher total selenium (> 0.300 mg/kg). However, the soils developed under the temperate (warm) steppe and desert conditions, e.g. chernozem, chestnut soil, calcic brown soil, desert soil and solonchak have moderate concentrations (0.140–0.300 mg/kg). The soils developed under the temperate (warm) humid/sub-humid conditions, such as brown earth, drab soil, dark brown soil and their similar types, like loessial soils, purple soil, red drab soil etc. have lower total selenium concentration. The purple soil is an exception. It is a lithogenic soil developed on the purplish rock formations low in selenium, even though it situates in subtropic zone. Originally the cultivated soil result is from different natural soil types by cultivation. Usually their wide range selenium concentrations are influenced both by the original soil selenium concentration and human activity.

3.2. Water-soluble selenium (WS-Se)

The geometric mean of water-soluble selenium concentration in all topsoil samples is 4.0 μg/kg and the arithmetic mean 6.4 μg/kg. The lowest is 0.6 μg/kg and the highest of 109.4 μg/kg. The water-soluble selenium concentrations (by geometric means, μg/kg) in different soils are in the following order:

Laterite (15.6) > solonchak (11.3) > chernozem (8.9) > desert soil (6.9) > yellow soil (6.3) > dryland cultivated soil (4.7) > meadow soil (4.5) = chestnut soil (4.5) > calcic brown soil (4.0) > grey forest soil (3.5) > red soil (3.4) > paddy soil (3.11) ≫ black soil (3.0) > swamp soil (2.4) > purple soil (2.0) > dark brown soil (1.9) = drab soil (1.9) > yellow brown soil (1.6) = brown earth (1.6) > red drab soil (1.1).

The soils developed under the humid tropic/subtropic and the temperate desert and steppe conditions have rather higher water-soluble Se concentrations, the soils developed under temperate humid/sub-humid conditions have lower levels of water-soluble selenium (< 3.0 μg/kg).

In general, soils developed under temperate humid/sub-humid conditions usually have low selenium levels both in total selenium and in water-soluble selenium.

3.3. Proportion of water-soluble Se to total Se in soil

The proportions of the two forms of selenium existing in soils are different. They can affect the Se-flux in geo-ecosystem and the health of humans and animals via the food chain. Thus it is very important to understand the proportion of the water-soluble selenium in soil for risk evaluation of soil Se influence on health. The percentages of water-soluble selenium to total selenium have been calculated and listed in Table 2 and the soil sequence based on the percentage (%, by geometric mean) shows as follows.

Solonchak (6.69) < desert soil (4.16) > chernozem

(3.99) > meadow soil ((3.57) > chestnut soil (3.06) > calcic brown soil (2.67) > purple soil (2.63) > dryland cultivated soil (2.47) > dark brown soil (2.04) > swamp soil (2.02) > drab soil (1.95) > grey forest soil (1.86) > yellow brown soil (1.80) > paddy soil (1.71) > laterite (1.65) > yellow soil (1.56) > brown earth (1.37) > black soil (1.28) > red soil (1.08) > red drab soil (1.07).
The percentages of water-soluble selenium to the total selenium decreased regularly from the soils developed in the northwest dry desert conditions to those developed under the southeast humid tropic/subtropic conditions. Generally the dry desert/steppe soil series have the highest, whereas the tropic/subtropic forest soil series such as laterite, red soil and yellow soil, etc., are the lowest. But the absolute water-soluble Se concentrations are still relatively high because of their higher total Se. The temperate forest soil series is situated in between.

3.4. Correlativity of water-soluble Se with total Se in soils

The correlation coefficient between the water-soluble Se and the total Se in soils is very significant in dryland cultivated soils (0.58; $P < 0.001$), whereas the lowest correlation coefficient was in paddy soil (0.23; $P > 0.05$) (Table 3). However, for natural soils developed under desert and steppe conditions such as desert soil, solonchak, chestnut soil and meadow soil, their correlations are either significant or very significant and in soils developed under forest conditions like red soil, brown earth and drab soil, the correlations are mostly insignificant.

3.5. Distributive patterns of soil selenium

The concentration of selenium in soil mainly depends on the two groups of factors. That is, geographically azonal factors such as parent rocks and landforms, etc., which determine the source of Se in soil and zonal factors such as biological and climatic conditions, etc., which influence or dominate the migration, maintenance, existing forms and availability of Se in soils to plants (Tan et al., 1994; Tan, 1996; Yang et al., 1990). Consequently, spatial distributive patterns of selenium are closely associated with geographical conditions. The present study indicates that the geographical features of the selenium in soils as following:

Table 3
Correlation coefficients between T-Se and WS-SE in soil

Soil type	N	r (coefficients)	P
Dryland cultivates soil	132	0.58	< 0.001
Paddy soil	39	0.23	N.S.
Red soil	10	0.51	N.S.
Yellow soil	9	0.68	< 0.05
Red drab soil	4	0.82	N.S.
Brown earth	4	0.58	N.S.
Gray forest	4	0.44	N.S.
Chestnut	11	0.66	< 0.005
Meadow soil	18	0.72	< 0.001
Solonchak	5	0.97	< 0.001
Desert soil	8	0.73	< 0.05

- The soils with low Se occur mainly in temperate humid/sub-humid geographical conditions, situated in the northeast to the southwest of China and forming a wide belt of low Se soil.
- On the two sides of this wide belt, the soils both in northwest and southeast of China belong to arid desert or steppe soils and humid tropic or subtropic soils respectively and they all have relatively high selenium concentration compared with the soils under temperate humid conditions.
- In the juvenile soils such as purple soil, the selenium contents are mainly dependent on the concentration of selenium in parent materials.
- There is relatively higher Se concentration in some large accumulation plains than those in the adjacent erosional areas.

The distributive laws mentioned above basically dominate the spatial distribution patterns and structures of soil selenium in China, forming a general scheme, shown in Fig. 1. It shows that the low Se soils mainly distribute in a broad belt in the middle of the country, running from the northeast to the southwest of China. In the northwest and the southeast there are two soil belts with relatively high selenium. However, the soil with abnormally high selenium contents mostly result from the parent rocks rich in selenium such as carbonaceous shale/sandstone. In our studies, four places of Se abnormality with topsoil of > 1.00 mg/kg have been recorded: Enshi in Hubei province, Ziyang in Shaanxi province and Zhijin and Jiuan (Guiyang city suburb) in Guizhou province.

Soil Se, which usually plays an important role in determining the Se concentration in food grains, vegetables, forages and even drinking water, is the basis of the Se cycle in geoecosystem. Through the ecological food chain, it influences human health in some areas of China, resulting in Se-responsive diseases such as Keshan disease and Kashin–Beck disease and endemic selenosis.

3.6. Selenium in soil and health

The effects of selenium deficiency and excess on animal health are well known in earlier Western literature. However, the direct effects on human health were not reported until early 1970s. When in China it has been observed that Keshan disease and Kashin–Beck disease are closely related to selenium deficiency in geoecosystems including soils, food grains, animal and human hair and human blood in disease-affected areas, compared with those in unaffected areas (EGAS, 1981, 1982; Tan et al., 1982, 1987a,b, 1994; Tan, 1989, 1996, Li, 1979, KDRG-CAMS, 1979). The Keshan disease is an endemic cardiomyopathy in man. The pathological changes associated with the Keshan disease mainly involve the myo-

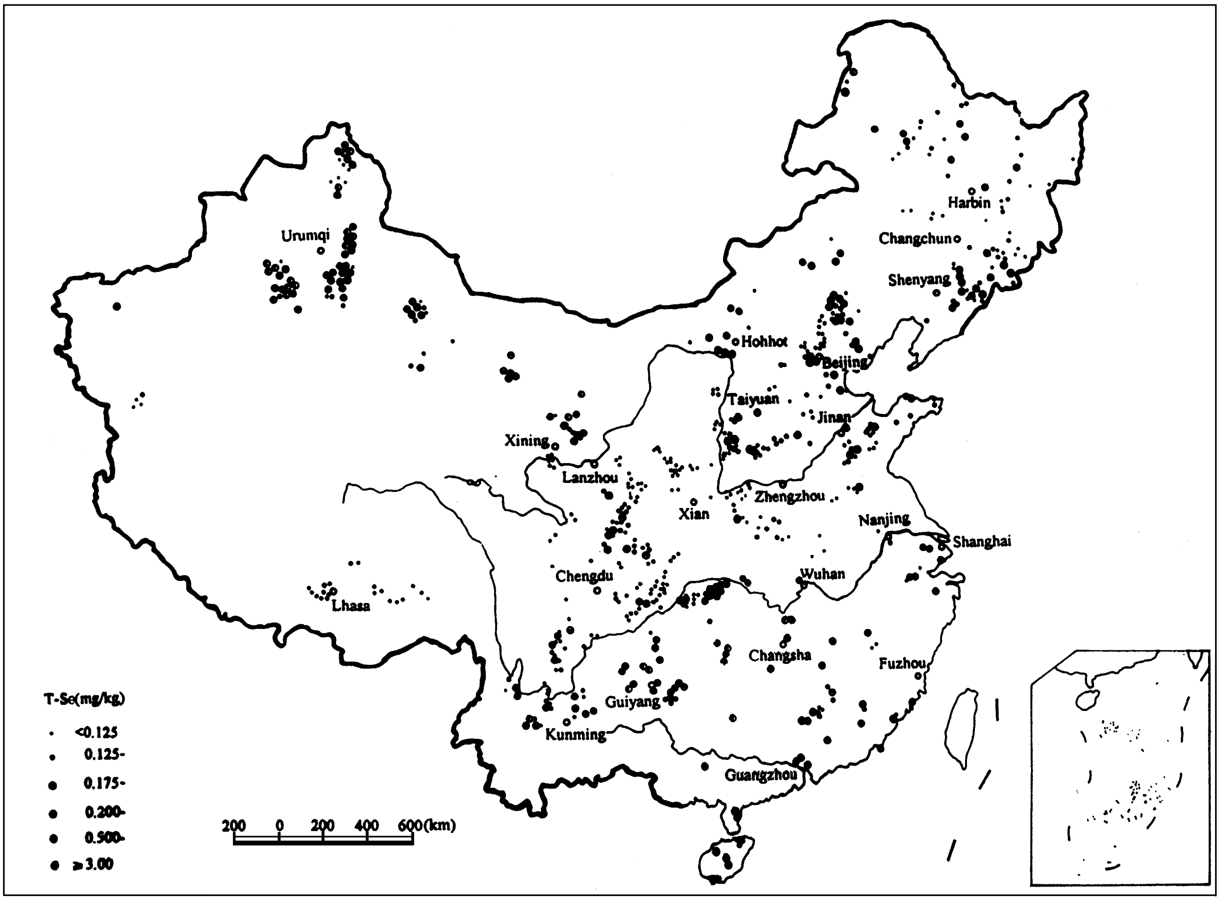

Fig. 1. Distribution of total selenium in Chinese soil.

cardium, serious myocardium degeneration, necrosis and scar formation. The principal clinical symptoms are acute and chronic cardiac insufficiency, acute cardiac failure, heart enlargement, gallop rhythm, electrocardiogram changes, etc. This disease has been found in 329 counties of 16 provinces, municipalities or autonomous regions, including Heilongjiang, Jilin, Liaoning, Inner Mongolia, Hebei, Shandong, Shanxi, Henan, Shaanxi, Gansu, Sichuan, Chongqing, Yunnan, Xizang (Tibet), Hubei and Guizhou in China (Tan, 1989, Fig. 2).

Kashin–Beck disease is an endemic osteoarthropathy. Pathological changes might be found in joints all over the body of the patients. But the major effects relate to joint cartilage and epiphyseal plate cartilage of the four limbs. The principal pathological changes are chondral degeneration and chondronecrosis. Its major clinical manifestations concern the joints of the four limbs. They become thickened, deformed and difficult to bend or stretch. Muscles become atrophied, the fingers, toes and limbs become shorter and a deformed body develops. Kashin–Beck disease, similar to Keshan disease, is distributed in 335 counties of 15 provinces, autonomous regions and municipalities of Heilongjiang, Jilin, Liaoning, Inner Mongolia, Hebei, Beijing, Shandong, Shanxi, Henan, Shaanxi, Gansu, Qinghai, Sichuan, Xizang (Tibet) and Taiwan (Tan, 1989, Fig. 2).

For a long time the cause of both the Keshan disease and Kashin-Beck disease were unknown. A variety of pathogenic hypotheses were reported, but with no satisfactory evidence. Since

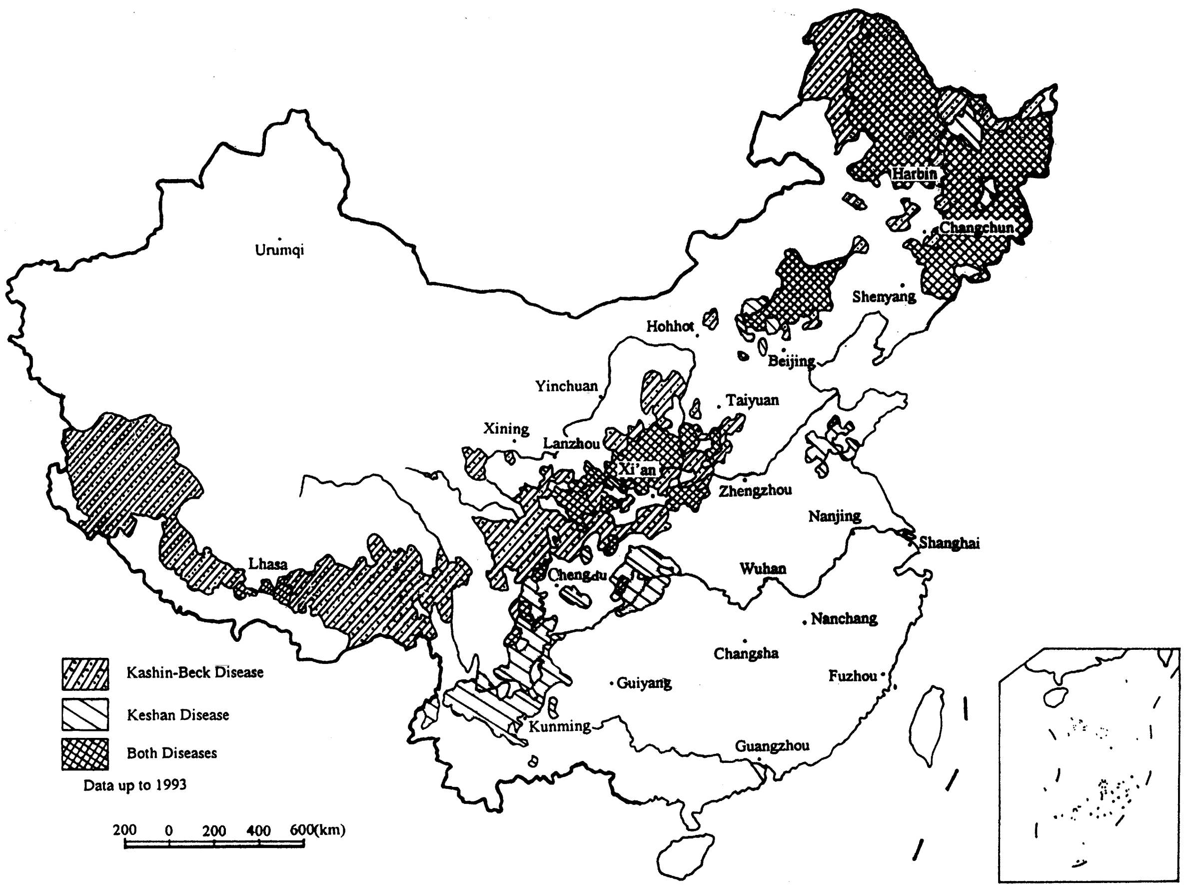

Fig. 2. Distribution of KSD and KBD (by disease-affected counties).

Table 4
Se contents of soils in the areas with and without KSD/KBD

Type	Area with KBD/KSD					Area without KBD/KSD				
	X	S.D.	G	S_{lg}	N	X	S.D.	G	S_{lg}	N
T-Se (mg/kg)										
Cultivated	0.112	0.057	0.100	0.1816	35	0.224	0.134	0.219	0.3297	161
Natural	0.119	0.075	0.105	0.2177	69	0.227	0.141	0.211	0.3192	86
WS-Se (μg/kg)										
Cultivated	2.5	1.0	2.5	0.3914	25	6.8	9.1	4.7	0.3106	151
Natural	2.8	2.2	2.2	0.2651	22	6.7	13.2	4.7	0.4452	71

the end of 1960s, extensive multidisciplinary investigations have resulted in an encouraging progress in pathogenic studies of the endemics. Two major findings supported the association between Se deficiency and the endemics and drawing a great interest among both the researchers at home and abroad: (1). The two diseases always occur in the area with low selenium eco-environment (EGAS, 1979, 1981, 1982; Tan et al., 1982, 1987a,b), (2). To prevent and treat the two diseases with Se achieved a great success (KDRG-CAMS, 1979, Li, 1979). Then the direct association of selenium to human health was first identified. In this context, soil Se is the basic factor for the occurrence of the two diseases. The relation of selenium in soil to the two endemic diseases is listed in Table 4.

In the KSD and KBD affected areas, the concentrations of T-Se and WS-Se both in cultivated and natural soils are lower than that in non-affected areas. The natural soils in affected areas usually include dark brown soil, brown earth, drab soil, yellow brown soil, red drab soil, loessial soil, purple soil and black soil. In disease free areas the soil types are mainly typical laterite, red soil, yellow soil, desert soil, chernozem, chestnut soil, calcic brown soil, etc.

The selenosis resulting from the excess of selenium in soil has been identified in Enshi of Hubei province (Yang et al., 1982, 1990), where the soil T-Se is up to 3.806 mg/kg, with an average of 2.314 mg/kg.

The distribution of topsoil Se in China and its relationship to Keshan disease and Kashin–Beck disease indicates that the total Se concentrations in topsoil of KSD/KBD affected areas are below 0.125 mg/kg. The marginal concentration is 0.123–0.175 mg/kg and the excessive level is more than 3 mg/kg. So these values could be considered as the reference threshold values for the risk assessment of soil potential selenium deficiency and soil Se excess.

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