Biogeochemical cycling of selenium in Chinese environments

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Abstract

The biogeochemical cycling and chemical transformation of Se in Chinese environments is reviewed, especially with regard to the formation of low-Se environments. The occurrence, distribution, concentration and flux of Se in Chinese environments are reported in comparison to those in the rest of the world. Low-Se regions in China can be classified into 4 categories. In the NE, organic matter in the soil affects bioavailability of Se in soil. In the Huabei plain, the concentration of soil Se is strongly influenced by leaching and hydrological transport processes and Se deficient regions are mostly located on the hill-slopes due to leaching. In the Loessial Plateau, low-Se soils have developed on the low-Se parent material with a very low flux of Se between soil and plants. Low-Se regions in the SW have soils derived from weathered materials of lower Se contents. Both the lowest and highest concentrations and the flux of Se in environments are reported for China. © 2001 Elsevier Science Ltd. All rights reserved.

1. Introduction

In China, research work on Se began during the 1960s. A major discovery from the intensive work, mostly geochemical surveys, was a low-Se belt, stretching from Heilongjiang Province in the NE to Yunnan Province in the SW. In the middle 1960s, research work began to focus on the association of the low-Se belt with endemic diseases, mostly the human Kaschin-Beck disease (KBD) and Keshan disease (KSD). In the 1970s, more epidemiological data was obtained, which showed a good correlation between the concentration of Se in the environment and incidences of KSD and KBD. These two diseases were later characterized as Se-responsive human diseases, which could be prevented by supplementing Se in a diet. Recently, research work has revealed the relationship between Se-deficiency and other endemic diseases, such as endemic liver cancer, endemic fluorosis, goitre and cretinism, etc. (Peng et al., 1995).

Among the comprehensive research papers on Se, many contribute to the environmental behavior, transport and chemical transformation of the element, as well as more profound thinking on the formation of low-Se environments. In this paper, the biogeochemical cycle of

Se in China, with special attention to the formation of the Chinese low-Se belt and the relevant geochemical processes are reviewed based on available data with a comparison to the global Se cycle.

2. Concentrations of Se in Chinese environments

The abundance of Se in the earth's crust is 0.05–0.09 mg/kg, about 1/6000 of the total S. However, the geographical distribution of Se is extremely variable. The total amount of Se in rocks comprises 40% of the total in the earth's crust, mainly in sandstone, quartzite and limestone. The worldwide Se content in eruptive rock is 0.01–7.0 mg/kg, 0.1–6.5 mg/kg in metamorphic rock, and 0.01–7.0 mg/kg in sedimentary rock (WHO, 1987). In China, the average abundance of Se in the earth's crust is 0.058 mg/kg and is lower than that in other parts of the world (Xia and Tang, 1990). The Se content of the various rock types is 0.07 mg/kg in metamorphic rocks, 0.067 mg/kg in eruptive rocks, and 0.047 mg/kg in sedimentary rocks, following the decreasing order of slate > clay rocks > basic and ultra-basic rocks > alkaline rocks > basalt > granite > hypersthene sandstone > limestone (Xia and Tang, 1990).

Se tends to accumulate in the carbonate fragments of sandstone, shale and slate, as well as phosphate-type rocks; rocks which occur in most of the Se-adequate

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regions. There are two notable Se-excessive or seleniferous regions in China, which are the Exi seleniferous region in Hubei Province and the Ziyang seleniferous region in Shanxi Province. The bedrock in the Exi seleniferous region consists of carbonaceous shale and high-Se coal.

The average concentration of Se in fresh water around the world is $0.2 \mu\text{g/l}$. The Se concentration in Chinese river water ranges from 0.04 to $5 \mu\text{g/l}$ and the soluble Se concentration ranges from 0.01 to $0.8 \mu\text{g/l}$ (Wang, 1991). In rural areas, such as in the Kaschin-Beck and Keshan disease region of the Chinese low-Se belt, Se concentration in well water can be as low as 0.11 ± 0.08 and $0.16 \pm 0.20 \mu\text{g/l}$, respectively (Wang, 1991). For most Chinese cities and in urban areas, the average Se concentration in drinking water is $0.65 \pm 1.06 \mu\text{g/l}$ (Wang, 1991). When a river is polluted, the soluble Se concentration can reach several mg/l (Li and Zheng, 1990). The highest Se concentration in drinking water has been reported in seleniferous regions. For example, the Se concentrations in two well water samples from the Exi seleniferous region were 8.4 and 72 mg/l , respectively (Wang, 1991).

The distribution of Se in soils is extremely uneven and significantly site-specific. Generally, Se-excessive districts are distributed in a spot-wise form, while low-Se regions are distributed in the form of a belt. Previous research has shown that the worldwide low-Se belts are

mainly situated along the 30° middle and high latitude in both the Northern and Southern Hemispheres. In China, a wide low-Se belt stretches from NE to SW, in the shape of a saddle, with a falling-off between 2 peak areas, including a semi-humid northeastern plain and a semi-arid Loess plateau, as well as most parts of the Tibet and part of the Yunnan Province (Fig. 1). On both sides of the low-Se belt, there are Se-adequate regions.

The major soil types in the low-Se belt are Brown Earth and Cinnamon soils, including 31 of the 41 total Chinese soil types (75.6%). The mean Se contents in soils in northwestern China (Se-adequate region), the middle part (low-Se belt), and the southeastern part (Se-adequate region) are 0.19 , 0.13 and 0.23 mg/kg , respectively (Tan, 1989). The Se content of soils depends on soil types, and soils with different Se contents can be classified into 4 categories as shown in Table 1.

Generally, vegetables contain less Se than cereals, the contents are in the range of <0.1 – 0.3 mg/kg in vegetables and are in the range of 0.02 – 0.8 mg/kg in cereals, in China. Significant differences can be found between plant concentrations of Se grown in Se-deficient and in Se-adequate regions. The difference is far more obvious than the difference of Se contents in soils of these regions (Table 2). The Se contents of cereals grown on Brown Earth and Cinnamon soils are higher than those grown on Red and Yellow Earth. The Se content of

Fig. 1. Geographic distribution of the Chinese Low-Se belt (source: Tan, 1989).

Table 1
Major soil types and their Se contents in China^a

Soil types	Se content (mg/kg)	Description
Steppy chemozems	≤0.1	Extremely low-Se
Mt. cinnamon soils		
Dark loessial soils		
Red brown earth		
Red cinnamon soils		
Subalpine meadows	0.1–0.2	Low-Se
NE Black soils		
Albic soils		
Dark brown earth		
Fluvo-aquic soils		
SE coastal paddy soils		
Chernozems		
Castanozems		
Brown pedocals		
Sierozems		
Desertous sand soils		
Castanozems-typed sand soils		
Purplish soils		
Yellow-brown earth		
Gray desert soils		
Brown desert soils		
Desert meadow soils		
Yangtze R. plain paddy soils	0.2–0.4	Se-adequate
Yellow earth zone's paddy soils		
Latosols		
Desert solonchaks		
Salinized meadow soils		
Phospho-calcic soils	> 0.4	High-Se
Limestone soils		
Yellow earth		
Red earth		
Lateritic red earth		

^a Data sources: Li and Zheng (1990) and Wang (1991).

cereals grown on desert soils and desert meadow soils is the lowest.

The Se contents of carnivorous foods are higher than in vegetables. They are in the range of 0.1–0.4 mg/kg. The Se content of the organs of animals and in seafood is much higher than in the ordinary foods consumed by human beings (0.3–0.6 mg/kg) (Wang, 1991).

The atmospheric Se in the poles and above the open sea, which could be regarded as an oceanic background value, is 0.04 ng/m³ (WHO, 1987). In the terrestrial environment, the reported atmospheric Se concentration varies in the range of ng/m³ to mg/m³ (Li and Zheng, 1990). The background in land atmospheric concentration of Se is lower than 1 ng/m³, as observed in the Nanjiabawa area (Tibet, China), but in a big city such as Beijing, it could be as high as 44 ng/m³ (0.7–44 ng/m³) (Li and Zheng, 1990). The major source of anthro-

Table 2
Mean Se contents in cereals in 3 typical regions of China^a

	Mean of Se content (mg/kg)		
	Northwest Se-adequate	Low-Se	Southeast Se-adequate
Rice	0.079	0.017	0.063
Wheat	0.087	0.018	0.052
Corn	0.050	0.015	0.048
Chinese sorghum	0.131	0.026	0.048
Millet	0.101	0.018	0.059
Highland barley	0.027	0.014	–
barley	0.028	–	–
Buckwheat	–	0.018	0.039
Soybean	0.120	0.019	0.088
Potato	0.033	0.018	–
Sweet potato	0.228	0.016	0.019

^a Data source: Li and Zheng (1990).

pogenic Se probably originates from the burning of fossil fuels. The Se content is in range of 0.04–10 mg/kg in coal and is in the range of 0.01–1.4 mg/kg in petroleum, in China (Wang, 1991). A higher content of Se was found in bituminous coal (5.7 mg/kg) and in brown coal (4.4 mg/kg), but it could be as high as 1524 and 23 mg/kg in coal and bone coal in the Exi seleniferous region (Wang, 1991).

3. The biogeochemistry of Se in Chinese environments

The biogeochemistry of Se is characterized by several distinct features, such as (1) the substitution of Se for S decreases if the oxygen fugacity increases; (2) the stable selenite are rapidly oxidized in surface conditions; (3) the adsorption of selenite on soil minerals is nearly quantitative. In different geochemical areas, Se behaves differently.

In northeastern China (Helongjiang, Jilin and Liaoning provinces; Fig. 1), the regional soils consist mainly of dark-brown earth (0.14–0.26 mg/kg) and black soils (0.13–0.39 mg/kg) in the temperate forest landscape zones (Yin and Ju, 1992). The content of organic matter in the soil is the highest in the country (6–10%), which immobilizes the Se in the soil. It was observed that Se tends to accumulate in the top layer. Both KBD and KSD have been reported where the soil Se content ranges from <0.1 to 0.3 mg/kg. However in regions where the soil Se content is higher than 0.2 mg/kg, and where KBD or KSD have been reported, the Se concentration in cereals was found to be very low. Se-deficiency relates closely to the Se bioavailability in the soil (Wang et al., 1991).

In the Huabei plain (including Hebei Province and Beijing; Fig. 1), the primary soils are Cinnamon and Fluvo-aquic, which exhibit basic and weak alkaline reactions. In such soils, the Se should be more mobile.

The biogeochemistry of Se is strongly dominated by the leaching and hydrological transport processes, resulting in the loss of Se in mountainous slopes (Hou, 1990). These processes are representative of the landscapes in the warm-temperate zones, including those found in Hebei, Shandong, Shanxi, Henan, and part of Qinghai provinces. Selenium deficient regions occur mostly on mountainous slopes and the KBD incidence and Se content of cereal, correlates well with the Se content of soil (Wang, 1991).

In the Loessial Plateau (Gansu and Shanxi provinces), low-Se regions are situated in semi-arid and semi-humid Temperate Zones and in the belt of the warm-to-temperate forests and grasslands (Fig. 1). The recent soils were developed from soils deposited during the Quaternary Period (0.044–0.066 mg Se/kg). The average Se content in loessial soil is 0.08 mg/kg, which is slightly higher than the parent's materials. In this area, soil Se content in the valley or river basin is higher than that in mesa or ridge, and is negatively correlated to annual rainfall (Jiang and Cheng, 1989). The Loessial Plateau in China is an important low-Se region where the Se-responsive endemic diseases generally prevail and are severe. The lowest Se flux in the world could be in this region (WHO, 1987).

Low-Se soils in Tibet are mainly alpine brown earth and alpine meadow soil, which exhibit weak acidic reactions. These soils undergo a very strong leaching process, where most of the soluble nutrients, including Se, are lost. In these areas, the region with lower Se is undergoing strong leaching, and the region with higher Se was formed during the orogenic movement. The soil Se content in several KBD and KSD regions is in the range 0.1–0.15 mg/kg.

The low-Se soils of southwestern China (Yunnan and Sichuan provinces) were those formed from weathered materials of sandstone and shale, especially from purplish sandstone and shale. In the same area, Se-adequate soils are of lateritic red earth found in the Southern Asia tropic zone, or formed from red limestone in the Central Asia tropic zone. Selenium contents in such soils show strong heritage from their parent materials (Xia and Tang, 1990).

On both sides of the low-Se belt, there are Se-adequate areas. The NW Se-adequate area is the most arid area in China, and is characterized by the lowest rainfall and the strongest evaporation. Soil types in the area are mainly desert and alpine soils. These soils are under strong oxidizing and alkaline conditions. In the basin and mountainous slopes, the soil Se has accumulated through the hydrological and aerial transport of fine particles and also by evaporation of the water. In these processes, Se is found in the form of selenate and is concentrated in the alluvial plain, the floodplain and the inland basin. In the surrounding area of the basin, which consists of alpine and sub-alpine soils, the soluble selenate reacts with organic matter and is retained in the soil.

In the Yangtze River and Zhujiang River drainage areas, the soil types are mainly Red and Yellow Earths and paddy soils. The red and yellow earths are high-Se soils (>0.4 mg Se/kg) and the paddy soil is a Se-adequate soil. The geochemical type for the paddy soil in the middle and lower reaches of the Yangtze River is acidic. The high content of Fe(III) and Al oxides favors Se accumulation in soil, in the form of selenite. In paddy soil, it switches from the oxidative to a reductive condition when irrigated, which leads to partial transformation of selenate to selenite, forming insoluble salts such as $\text{Fe}_2(\text{SeO}_3)_3$ and $\text{Fe}_2(\text{OH})_4\text{SeO}_3$. Consequently, the Se content is lower in the topsoil, and higher at depth.

Se-excessive soil in the Exi seleniferous region was developed from carbonaceous shale and outcrops of high-Se coal, and maybe more important in the slopes of the Qing River basin. The high-Se coal was actually formed from an ancient forest (Yan, 1993).

4. The mobility and bioavailability of Se in Chinese environments

It is generally recognized that the uptake of Se by plants may not necessarily depend on the Se content of the soils. For example, soils of Hawaii containing 6–15 mg/kg Se do not produce seleniferous (toxic) vegetation, while soils of South Dakota and Kansas containing <1 mg/kg of Se produce toxic vegetation (Meyer and Burau, 1995). It has been reported that plant Se concentration depends significantly on the soluble fraction of soil Se, instead of the total Se in the soil. However, the soluble fraction of soil Se is a function of soil physico-chemical properties and varies with biological reactions.

Simulation experiments (Wang, 1990) has shown that the maximum adsorption capacity of Se on Chinese soils followed the order of: gray desert soils $<$ Castanozems $<$ dark Brown Earth $<$ Chernozems $<$ Black soils, which is the order of increasing Se content in the soils. The mobility of soil Se is mainly controlled by Fe(III), Mn and Al oxides in moist and semi-moist acidic soils and by Ca, Mg, and K oxides in semi-dry alkaline soils (Hou et al., 1991). Within soil components, the adsorption capacity of Fe(III), Mn and Al oxides are larger than those of clay minerals (Xu, 1988). Consequently, the soluble Se concentration of the main soil types decreases, following the order of ferralsols $>$ ferrisols $>$ luvisols $>$ isohumisols $>$ aridisols (Wu et al., 1997). Beside minerals, a positive correlation between contents of soil Se and organic matter was observed in NE China (Zhang, 1990).

The transport of Se from soils to plants is affected by soil pH, Eh and co-existing anions, as well as dissolved organic matter in soil solutions (Wang et al., 1996). For example, organic matter or humic substances may carry Se in their matrix; more than 90% of the total Se in the

soil solution was found in the organic fraction (Wang and Peng, 1988).

Selenium concentration in soil solutions may be a better indicator for defining Se deficiency than the soil total Se. In low-Se soils, the soluble Se concentration could be as low as 2 µg/kg soil or about 1–2% of the soil Se. In Se-adequate soils, soluble Se constitutes about 4–6% of soil Se or as high as 18 µg/kg soil. There is a correlation between the concentration of Se in soil solution and the Se concentration in corn, wheat and rice grown in Shanxi Province ($r=0.810$, 0.727 and 0.723 , respectively). In NE China, the observed correlation coefficients between the concentration of Se in soil solution and the Se content in soybean, corn and rice were 0.995 , 0.995 and 0.996 , respectively (Wang, 1991).

Only some of the Se found in soil solutions is bio-available to plants. Generally, 45% of soluble Se in soil solutions can be utilized by plants or about 0.5–1.0% of soil Se in low-Se soils is available for plant uptake. In an in situ experiment, using ^{75}Se as a tracer, it was estimated that about 0.2–1.3% of soil Se can be available for corn grown in the soils of the NE China (Li and Zheng, 1990).

The partitioning of Se, in different part of plants, decreases following the order of root > stem > leaf > seed in cereal crops. In an in-situ spiked ^{75}Se experiment, it was shown that there was a decreasing order of Se in granule > Se in the main part of the leaf > Se in root > Se in seed shell > Se in tip of leaf > Se in stem (Li and Zheng, 1990).

5. Se flux in global and Chinese environment

It has been calculated that the global ocean-to-atmosphere Se flux is $5\text{--}8\times 10^6$ kg of Se/a, part of which can be brought back to the terrestrial environment (WHO, 1987). This flux is about two thirds of the global natural flux of Se to the atmosphere ($6\text{--}13\times 10^6$ kg), and is much higher than the annual industrial Se production in the world (1.4×10^6 kg). The anthropogenic contribution of Se flux to the atmosphere is about 30–50% of the total, coming mainly from coal combustion, nonferrous metal production, smelting, refining and fertilizers. The total emissions amounted to 3.6×10^6 kg in the United States, while it was only 0.028×10^6 kg in Finland (Conde and Alaejos, 1997). Through the combustion of fossil fuel, human activity emits an annual $2.5\text{--}4\times 10^6$ kg of Se in the world. Based on annual coal production ($0.8\text{--}1.2\times 10^{12}$ kg), the mean Se content of coal (1 mg/kg) and the of gaseous Se emitted during combustion (50–90%), emission of Se from coal combustion should be about $0.4\text{--}1.1\times 10^6$ kg of Se/a in China. Emission through coal combustion becomes the major pathway of Se in the global cycle.

The mean Se content in 8 ice cores from Greenland over the period of 800 BC to 1968 was 0.018 µg Se/L

(WHO, 1987). Therefore the estimated Se flux through rainfall (500–1000 mm) should be 0.09–0.18 g Se per ha. The concentration of Se in inland rainwater of China is in the range of 0.03–0.59 µg Se/L, with the highest frequency concentration in the range of 0.1–0.2 µg Se/L (Zhu and Tang, 1988). Rainwater with a Se concentration of 0.03 µg Se/L was observed over several rainfalls and can be taken as clean inland rainwater. The value of 0.03 µg Se/L was the closest value to the Se concentration in Greenland ice samples (WHO, 1987). Accordingly, the annual flux of Se through rainfall was about 0.15–0.3 g per ha or $0.6\text{--}1.2\times 10^6$ kg of Se/a in China. Generally, the Se content of rainwater in coastal areas is lower than that of interior landscapes, probably because of the influence of oceanic transport.

Se flux from dry precipitation (dust-fall) was strictly due to the pick up of local dust and, therefore, was positively correlated to the Se content in local rainfall (Zhu and Tang, 1988). Higher Se content (0.5–5.0 mg/kg) was observed in air-borne particles than in surface soils, because of its accumulation on fine particles. The long-distance transport of Se through air-borne particles to the low-Se belt should not be a major pathway for the Se cycle.

According to simulation experiments, the annual flux of Se leaching loss per hectare of agricultural land in the low-Se belt should be 2–4 g/a (Wang, 1991). An equivalent calculation of the leaching loss can be made, based on an assumption that 100% of water soluble Se could be leached during the rainy season in a year. Because the water soluble Se (or Se in soil water) represents 1–2% of the total Se in soils with Se contents in the range 0.08–0.12 mg/kg, it results in a maximum loss of 2–8 g/a per hectare. The estimation based on the leaching experiment was in good agreement with that based on the leaching of soluble soil Se. Selenium leaching loss from terrestrial to aquatic environments varies with environmental conditions. For example, while the soluble Se concentration in the soils of NW China is higher, the annual leaching loss of Se per ha may not be as high as that in the low-Se belt, because of significantly lower rainfall (<400 mm). In SE China, the flux of Se between terrestrial and aquatic environments is limited by the fixation of Se on Fe and Mn oxides.

In ^{75}Se labeling experiments, the rates of volatilization of Se from different types of soils were between 0.001 and 0.092% of soil Se and did not correlate with the Se content of soils (Wang, 1990). In laboratory simulations without addition of Se, the rates of evolved Se from low-Se soils were between 0.1 and 0.27 ng/kg/d when incubated under room temperature for 17 days (Wang et al., 1989). The above 2 estimations are actually comparable, and the estimated maximum annual net loss from volatilization should be 0.08–0.20 g/ha or 0.001–0.092% of the total Se/a in different soil types. This leads to a net loss of $0.011\text{--}0.027\times 10^6$ kg from

agricultural land in China (not including grassland and forest). Comparing this flux with the flux through the leaching process, the volatilization loss of Se is negligible.

The partitioning coefficients of Se between soil and different parts of wheat were obtained by using an in-situ ^{75}Se tracing approach in NE China (Li and Zheng, 1990). Within the plant and among the above ground parts of wheat, the seed contained 59–63% of the radioactivity, of which 68–72% was found in the flour. Accordingly, plantation in NE China removes a total of 0.05–0.20 g Se from 1 ha soil/a, in which only 0.03–0.12 g Se was harvested as seed. The rest of plant was left on the ground or burned as fuel. Therefore the total flux in 1 ha of land through plantation, and plant utilization, should be between 0.05–0.20 g or $0.007\text{--}0.028 \times 10^6$ kg/a in China. The flux of Se through plantations and harvests can be estimated by using data for the Se content of cereal, and mass transfer. The net loss of Se from agricultural land due to harvestation is $0.009\text{--}0.036 \times 10^6$ kg/a, according to a national crop production of 0.48×10^{12} kg/a. The 2 estimations yield very similar results. Generally, the Se content of the cereal in the NW China is 4–5 times as high as that in the low-Se belt, while the Se content of the cereal in the SE China is 2–4 times as high as that in the low-Se belt. The flux of Se in a low-Se soil/plant system is illustrated in Table 3.

Zhang (1992) have estimated the worldwide Se flux through vegetation. The annual flux caused by primary production is 2–3 g per ha in tropical forest, 0.25–0.65 g per ha in temperate forests, and 0.3–1.1 g per ha in temperate grasslands. In desert areas, there is only an annual of 0.07–0.18 g of Se per hectare in the cycle. The Se flux in the low-Se belt of China is similar to that in temperate grasslands.

The worldwide annual weathering flux of Se was estimated to be 0.2×10^6 kg (WHO, 1987). In comparison, an annual Se consumption through food or food products is 0.1×10^6 kg, which should not cause a net loss of Se from the terrestrial ecosystem. Net loss from the terrestrial ecosystem includes flux between the terrain and the atmosphere, and flux between inland waters and oceans. The latter was estimated to be 7.4×10^6 kg/a in the world. It could be concluded that the major process

causing the loss of Se from the terrestrial environment should be through leaching, while the major process causing the gain of Se in the terrestrial environment should be due to fossil fuel combustion. In recent years, more and more farmers in Chinese low-Se areas use coal as their new energy source, and use increasing amounts of fertilizer in their agricultural activities. It is anticipated, therefore, that an increase of Se flux in local environments will occur, which has a positive role in the amelioration of Se health status.

6. Recommendations for future work

Se behavior in the geosphere and its function in the biosphere are very specific. Although a relatively large database already exists, Se biogeochemistry and cycling needs far more research, especially under human stress. In addition, the narrow margin between beneficial and harmful levels has important implications for human activities that increase the amount of Se in the environment. The increase of Se concentration in the environment may improve the current situation of Se-responsive diseases or may cause environmental pollution, due to its bioaccumulation in wild animals or humans (Lemly, 1997). Therefore, it is necessary to monitor the changes of the various fluxes of Se in the environment in the future.

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Table 3
Flux of Se in a low-Se soil/plant system in China (g Se/ha/a)

Process	Annual net loss	Annual net gain
Soil inventory	(188–375)	
Volatilization	0.08–0.20	
Rainfall		0.15–0.30
Agricultural fertilizer		2–4
Leaching	2–4	
Plant uptake	0.03–0.12	
Balance	(–)2.11–4.32	(+)2.15–4.30

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