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Constraint on selenium bioavailability caused by its geochemical behavior in typical Kaschin–Beck disease areas in Aba, Sichuan Province of China

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HIGHLIGHTS

- Inheritance from Se-deficient parent materials is predominant in typical KBD areas.
- The water-soluble Se with a negative correlation to KBD plays a pivotal role.
- Behaviors for Se(VI) and Se(IV) in various conditions influence Se bioavailability.
- Diet structure is an additional constraint to local human daily Se intake.

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ABSTRACT

Kaschin–Beck disease (KBD), an endemic osteoarthropathy, is distributed in the low-selenium (Se)-belt that stretches from northeast to southwest China. However, very few studies have investigated the relationship between low bioavailabitity of Se and KBD. The present study examined the behavior of Se and other elements in areas with varying levels of KBD prevalence using pedological and geochemical methods. Rhizosphere soil samples obtained from the KBD-stricken Aba area were classified into Ustic Isohumisols (J2), Udic Luvisols (L4), Stagnic Gleysols (I2), and Cryic Cambisols (M1) and the integrated constraints on selenium bioavailability in these soils were analyzed. We found that Se concentration in soil profiles from a typical KBD area ranged between 0.08 $\mu g \cdot g^{-1}$ and 0.215 $\mu g \cdot g^{-1}$, indicating absent and marginal bioavailability, respectively. This suggested that low Se bioavailability may be a feature that soils inherit from their Se-deficient parent materials. Moreover, the soil types examined showed different geochemical behaviors such as eluviation for soluble Se(VI), migration of Se(IV) for its adsorption on clay and sesquioxide, and extreme redox conditions. In conclusion, a higher level of Se bioavailability in environment might be related to a lower risk of KBD, and our results offer a foundation for scientific theory on ecological geochemistry and improve our understanding of KBD.

1. Introduction

Kaschin–Beck disease (KBD), an endemic osteoarthropathy characterized by deformity of affected joint cartilage and epiphyseal plate cartilage of the four limbs, poses a serious threat to quality of life (Tan and Huang, 1991; Wang and Xie, 2005) by causing severe pain and discomfort, limited mobility, anxiety and depression (Farooq et al., 2011), and shortening of lifespan (Foster and Zhang, 1995). KBD afflicts at least 2.5 million people in China and neighboring parts of Russia and North Korea (Stone, 2009). It is primarily distributed in the low-selenium (Se)-belt that stretches from northeast to southwest China in the shape of a large saddle (Tan et al., 2002; Zhang et al., 2010). Elucidation of the relationship between KBD and low Se has led to growing appreciation of the importance of Se to human health. Se is an essential component of the biological enzyme glutathione peroxidase (GSH-Px), which acts as an antioxidant to prevent tissue degeneration (Navarro-Alarcón and López-Martínez, 2000; Keshavarzi et al., 2012). However, it is difficult to maintain a safe level of Se intake in adults, owing mainly to the relatively narrow gap between Se-deficiency (<40 $\mu g \cdot day^{-1}$) (Levander, 1997) and excessive Se dose (>400 $\mu g \cdot day^{-1}$) (Navarro-Alarcón and Cabrera-Vique, 2008). As a result, many diseases such as Keshan disease (cardiomyopathy), cancer, hypertension, and selenosis commonly occur in addition to KBD.

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Moreover, mathematical manifold models, such as that showing the relationship between prevalence of KBD (y) and concentration of Se (ρ), and illustrated by the equation $y = 0.00016\rho(Se)^{-2.42}$, ($\gamma = -0.98$, $\gamma_{0.01} = 0.71$), also exist (Cao et al., 2004). Although the etiology of KBD remains under debate, various hypotheses showed a correlation with alimentary myco-toxicosis (T-2 toxin) (Yang, 1995), humic substances (humic acid or fulvic acid) in drinking water (Peng et al., 1999), and parvovirus B_{19} virus infection (Wang, 2005). However, some evidence suggested that Se deficiency could simply act as a risk for KBD, and that the actual disease is caused by oxidative stress (Suetens et al., 2001).

From the view of medical geology, the medically important element Se is influenced by the rock–soil–water–plant–animal/human geochemical cycle (Dissanayake and Chandrajith, 2009). For the topsoil system, a variety of input pathways such as precipitation, irrigation, and fertilizer and output pathways such as infiltration, crop harvesting, removal of straw, and Se volatilization have been analyzed to estimate the proportion of human and natural influence (Yu et al., 2014). On a worldwide scale, Se distribution has been explored with particular focus on natural global cycle of oxidation and reduction processes of Se species among the terrestrial environment, marine environments, and the atmosphere (Winkle et al., 2012). Although relocation of residents from areas with special geological environment is often done to reduce the occurrence of KBD, the exorbitant cost involved makes this measure impractical and the pathogeny of KBD remains controversial (Stone, 2009). Changes in topographical features and leaching/erosion processes control the migration of Se into soils (Fernández-Martínez and Charlet, 2009). However, the importance of its bioavailability depends on speciation, ionic strength, pH or redox potential, and in particular, crop Se uptake (Fairweather-Tait et al., 2011). Lv et al. (2012) investigated the distribution of Se in rocks, soils, drinking water, and crops and their relevant bioavailability in typical KBD areas, and found that KBD is prevalent and remains uncontrolled under conditions of low-bioavailability of Se. Thus far, new KBD cases have been reported mainly in the Aba autonomous prefecture of Sichuan Province, the Tibet Autonomous Region, and the southern region of Qinghai Province after integrated prevention measures were taken in China (Zhang et al., 2011). In the present study, we take the Aba area, located in the eastern edge of the Tibetan Plateau, as an example of a typical KBD area.

To explore the relationship between KBD and Se, one must first consider the fact that there is no significant difference in Se levels between KBD patients and control individuals from the same area (Li et al., 2012). This may be a challenge to our research. Notably, strong evidence supporting a link between KBD incidence and Se-deficient environments did not exist until recently. The complex dietary structure of animals and humans enables foods such as organ and muscle meats and cereals, which are major sources of Se, to deliver necessary nutrients more effectively than drinking water (Appleton et al., 2006). In the present study, we used a combination of pedological and geochemical methods to examine soil profiles, crops, rhizosphere soils, and human hair, blood, and urine samples obtained from the Aba area, in order to determine the integrated constraints on Se in discrepant KBD-affected regions. Our results offer a scientific theory on ecological geochemistry of KBD and could be beneficial for the prevention of this disease.

2. Materials and methods

2.1. Study area

The Tibetan Autonomous Prefecture of Aba is located in the northwestern region of Sichuan Province in China (Fig. 1b). The clinical prevalence rate of each county determined on the basis of the diagnostic criteria of KBD (GB16003-1995) proposed by the Ministry of Health of the People's Republic of China is shown in Fig. 1b. We selected typical KBD counties, including Zamtang, Aba, Jinchuan, Barkam, and Hongyuan (Fig. 1a), in which isolated pockets of KBD are found, as study areas. Quaternary unconsolidated formation in the Aba area mainly includes alluvium, eluvium, deluvium, and some glacier sediments, and the majority of their components are siallites, followed by carbonates. According to Chinese Soil Taxonomy (CST), these deposits are categorized into four suborders, namely Stagnic Gleysols (I2), Ustic Isohumisols (J2), Cryic Cambisols (M1), and Udic Luvisols (L4) (Fig. 1a; Gong, 1999). The climates of these regions underscore various topographical features. Weather in the rough plateau landscape is the same as that of a typical continental plateau, featuring slight differences in the temperature of its four seasons; on the other hand, the Alpine plateau landscape with a gorge has a cool subhumid climate and strong distinction between dry and wet seasons. The statistics describing different degrees of KBD-stricken areas are shown in Table 1.

2.2. Sampling and preparation

The field work was conducted in the summer, which is a slow farming season, from 2009 to 2011. Based on the degree of KBD and topographical features, soil profiles, crops, and their rhizosphere soils, as well as hair, urine, and blood samples from inhabitants were collected from typical affected and control areas mainly distributed in two geomorphic units including a rough plateau and an Alpine plateau with a gorge. Some areas were located on hillsides, while others were in valleys (Fig. 1a and Table 1).

The majority of soil profiles were collected at an altitude of 3000 m above sea level, dug until bedrock was exposed. Each soil sample was $40 \times 20 \times 2$ cm in volume and weighed more than 1 kg, and was collected every 2 cm from top to bottom on the basis of measurement by a wooden level staff to indicate the history of soil profile generation. To ensure the accuracy of sample density, we used a plumb and a leveling rod with a bubble for vertical and horizontal measurements, respectively. In Aba, a typical KBD area, 28 soil profiles were sampled away from roads, garbage dumps, and other obvious areas of human contamination. Five additional soil profiles were collected in Qinghai Province at a sample density of 1 sample/10 cm for comparison. Each sample was placed into a cotton ordinal bag with a bamboo spade and was screened through a 20-mesh (<0.84 mm) nylon sieve after being dried thoroughly in the shade. More than 100 g of the sieved soil particles was then stored in a clean #6 zipper-lock bag (10×14 cm), and carried to the laboratory for chemical analysis. The remaining samples were stored in a #10 zipper-lock bag (24×34 cm) as duplicates. Prior to analysis, the soil samples were dried at a maximum of 40 °C to avoid loss of volatile elements and were ground into powder (<0.074 mm) using high-alumina agate mills (200-mesh).

A total of 139 samples of rhizosphere soil, which surround the roots of plants such as highland barley, potatoes, maize, and other vegetables, were collected from a typical KBD area in Aba, and 20 rhizosphere soils were collected from vegetable gardens in KBD-free areas of Yanting County for comparison. After removing plant residues and stones, samples of rhizosphere soils were prepared by the same method used for soil profiles, as described above. Moreover, an additional bag containing more than 100 g of 20-mesh sieved soils was sent to the Key Laboratory of Chemical Synthesis and Pollution Control at China West Normal University in Nanchong, Sichuan Province, China, to determine total Se concentration and dissolved Se species. Prior to analysis, soil samples were dried in an oven at 80 °C for 24 h, and were then transferred to a mortar, crushed, and screened through a 100-mesh (<0.15 mm) nylon sieve. The 100 g-weight sieved soils were transferred to 1000 ml test tubes, shaken and extracted 5 min after 500 ml deionized water was added. The soil solution was allowed to stand for 4 h and was then filtered through a 0.45 μm aqueous membrane by vacuum filtration.

To explore the correlation between plants and their corresponding rhizosphere soils, we collected 18 samples of ripe barley kernels and 33 samples of potatoes, which are major components of the local diet. These perfectly matched the sampling sites of the 139 rhizosphere soil samples. Each 250 g sample contained a mixture of soils from four