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Selenate redistribution during aging in different Chinese soils and the dominant influential factors

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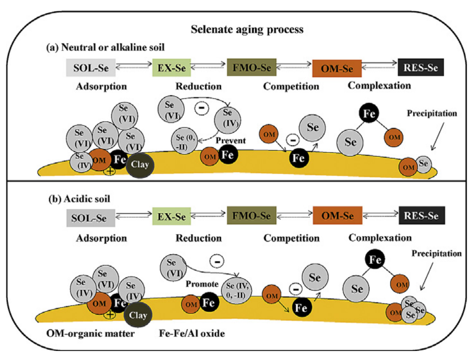
HIGHLIGHTS

- The speciation transformation of Se is differed greatly with soil pH during one year aging process.
- The dynamic changes of soil available Se content could be well described by Elovich model.
- Several decades were needed for exogenous selenate to reach natural available Se distribution.
- Soil pH, amorphous Fe and Al oxides and organic matter dominantly influenced selenate aging process.

GRAPHICAL ABSTRACT



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ABSTRACT

To date, few works have attempted to determine the effect of soil types on Selenium aging process and the possible influential factors. In this study, the differences in Se speciation distribution and availability in 15 Chinese typical agricultural soils were investigated using spiked selenate for the entire year. Results evidenced that after one year of incubation, Se transformed from soluble fraction to Fe/Mn oxides and organic matter bound fractions in neutral or alkaline soils (pH 7.09–8.51) and from exchangeable fraction to residual fraction in acidic soils (pH 4.89–6.82). The available Se content in all soils declined rapidly at the initial stage of aging, with most of the neutral or alkaline soils reaching equilibrium after 109 d, whereas the acidic soils reached equilibrium after only 33–56 d. The available Se content in soil decreased constantly during the entire aging process in S4 (Xinjiang Gray desert soil), S12 (Anhui Yellow brown earths), and S15 (Hunan Krasnozems). Elovich model was the best model ($R^2 > 0.80$) in describing the Se aging process. Estimated time for exogenous Se reaching the distribution of available Se in corresponding native soils extended from 9.7 y to 50.2 y, indicating a much longer time was required for spiked soil to reach equilibrium. Soil pH was the most significant factor directly and negatively influencing the aging process ($p < 0.05$), while organic matter played a dual role on Se speciation. Results could provide reference for the selection of unified equilibrium time on Se-spiked experiment.

1. Introduction

Selenium (Se) is an essential trace element for human and animals. It is a "double-edged sword" element that may negatively

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affect the human body when inadequately or excessively taken (Hartikainen, 2005; Mombo et al., 2015). Approximately 0.5–1 billion of the population have suboptimal Se intake (Li et al., 2010) with an increased risk of several diseases (Combs, 2001; Han et al., 2013). In addition, with a narrow range among deficiency, adequacy, and toxicity, safely and effectively supplying Se to human and animals becomes a study hotspot for multiple disciplines. The current generally accepted secure method to increase Se level in human body is biofortification through soil–plant–animal (human) transportation (Chilimba et al., 2012; Premarathna et al., 2010, 2012). Soil fertilization with selenate is one of the most common biofortification strategies successfully applied in some western countries such as Finland and Britain (Broadley et al., 2007). Some studies have been conducted with exogenous Se application for Se environmental risk assessment and fertilization recommendations, including field simulation experiments (Fan et al., 2015; Jiang et al., 2015), pot experiments (Li et al., 2016; Peng et al., 2016), adsorption and desorption tests (Feng et al., 2016; Li et al., 2015b), leaching experiments (Schwartz et al., 2016), and aging incubation (Di Tullo et al., 2016; Li et al., 2016; Tolu et al., 2014a).

When exogenous Se is applied to soil, it experiences several complicated reactions, such as solid–liquid distribution including complexation, surface adsorption, ligand exchange, chelation, precipitation, and micropore diffusion. Its extractability, bioavailability, or toxicity decreases with time extension, which is called aging or fixation (Jalali and Khanlari, 2008; Wang et al., 2015; Zheng et al., 2013). The solid–liquid distribution (mainly adsorption) is the dominant mechanism for short term aging process, influenced by soil colloid particle structure, pH, and redox conditions (Wang et al., 2015). Long-term aging process is predominantly controlled by micropore diffusion, involving ligand exchange, mineralization, fixation, and so on, and affected by time and environmental factors. The longer Se exists in soil, the greater its availability is degraded and becomes more stable (Di Tullo et al., 2016; Li et al., 2016; Tolu et al., 2014a). Anthropogenic input metals are much reliable and bioavailable than metals in native soil (Liu et al., 2015; Ma et al., 2015); hence, this discrepancy is always ignored for a long time. Recognizing and predicting the exogenous Se aging process for environmental risk assessment and soil quality evaluation is important because aging plays an important role in altering soil metal availability and toxicity. In addition, researchers found that the Se aging rate or bioavailability is not only dependent on total Se content but also more importantly on Se speciation in soils (Fordyce, 2013; Wang and Chen, 2003; Wang et al., 2012). The importance of aging, which represented the process of speciation and redistribution of Se in soils, should be highly concerned with the labile fractionations being transformed into steady fractionations (Keskinen et al., 2011; Li et al., 2016; Tolu et al., 2014a).

To date, most of the aging studies on Se are focused on selenite speciation given that it is much preferable for soil adsorption. Tolu et al. (2014a) and Di Tullo et al. (2016) used isotope ^{77}Se (IV) for short- (3 months) and long-term (2 years) Se mobility investigations and found that exogenous selenite is mainly contributed to soluble and exchangeable fractions and transformed to stable fractions in agricultural soil during aging. On the contrary, the organic-bound Se content increased, and the soluble fraction reached equilibrium after 1–6 months. Li et al. (2016) studied the performance of exogenous selenite in three different types of soil for 100 d and concluded that Se aging rapidly declined initially and slowly decreased after the process. Combined with biological test, they recommended a 21–30 d equilibrium period before the Se reaches stabilization. However, studies on the aging of selenate in soils were still limited on short-term adsorption experiments and relied on solid–liquid distribution coefficient (K_d) (Bruggeman et al., 2007; Feng et al., 2016); long term aging is rarely reported.

The Se speciation in soil is basically controlled by three mechanisms: oxidation vs. reduction, mineralization vs. mobilization, and volatilization (Ros et al., 2016; Sharma et al., 2015), which was controlled by soil properties such as pH, organic matter, iron/aluminum/manganese oxides, and clay content (Coppin et al., 2006; Li et al., 2016). Meanwhile, the interaction between exogenous Se and soil components, such as adsorption/desorption, dissolution, and precipitation, and redox reaction and ligand determine the availability. Soil pH is a key factor that reflects Se bioavailability because it is strongly immobilized in acid and reductive soils (Ghosh and Singh, 2005; Sharma et al., 2015). The iron/manganese oxides were the most powerful adsorbents for Se, followed by organic matter; clay was the least powerful (Zhang et al., 2012). Se dissolution happened under reduced condition with dissolved iron/manganese valance oxide release and soluble organic matter fulvic acid bound Se precipitation (Keskinen et al., 2011). In addition, biogeochemical factors (humidity and temperature) and soil physicochemical properties could also affect aging process (Duan et al., 2014; Takeda et al., 2013). Many studies have illustrated the effects of soil properties on Se redistribution. However, such studies have been conducted on either individual soil type (Dhillon et al., 2007; Han et al., 2013; Li et al., 2015a) or focused on 2–3 soil types (Di Tullo et al., 2016; Tolu et al., 2014a). The information on the influence of different soil properties on the speciation and redistribution of Se during the aging process is rarely available. Consequently, in the present study, 15 different types of soil from different Chinese provinces with different soil physicochemical properties were selected and spiked with selenate to investigate the speciation distribution of Se during 364 d incubation period. This study aimed to (1) examine the aging effect on spiked selenate in terms of speciation and availability in different soil types; (2) better understand the relationship between aging equilibrium and soil physicochemical properties, or dominant factors that affect aging; (3) provide reference for reasonable application of selenate in biofortification and environmental risk assessment.

2. Materials and methods

2.1. Soil samples and their physiochemical properties

Soil samples were collected from farmlands of 15 main soil types, which are distributed in 15 provinces in China (Fig. 1). Samples were obtained from the surface layer (0–20 cm depth) with great distinct physicochemical properties, due to parent materials, climate condition, and crop system differences (Table 1). Soil

Fig. 1. The distribution map of sampling soils (S1~S15 are represented for each soil type sampled from different provinces in China).