

Review Articles

Application of Rare-earth Elements in the Agriculture of China and its Environmental Behavior in Soil*

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Abstract. Rare-earth elements (REEs) have been used in fertilizers in the agriculture of China for about 20 years. They have been shown to be beneficial elements for plants. For example, they have improved the yield and quality for several kinds of crops. This paper reviews the current literature on studies of REEs being used as fertilizers. Some studies have focused on the effects of REEs on metabolic nutrients, photosynthesis and stress resistance of plants. Other studies have shown that the environmental behaviors of REEs in soil are dominated by their low solubility. Fluorides, carbonates, phosphates and hydroxides may form neutral complexes containing REEs with a low solubility. The amount of extraneous REEs demonstrate the following relationship: residual >> bound to organic matter > bound to Fe-Mn oxides > bound to carbonate >> exchangeable and water soluble forms. The adsorption capacity of REEs depends on the clay type and the content of amorphous and manganese oxides, whereas the desorption of REEs is usually very low. At the end of the paper, authors discussed the needs for future environmental research on REEs, which would shed new light on the effects of REEs on agriculture, environment and human health.

Keywords: Agriculture; China; rare-earth elements (REEs); soil

Introduction

The rare-earth elements (REEs) consist of 15 elements with successive atomic numbers (Z from 57 to 71, and Sc, Y), and similar chemical and physical characters. One of them, Pm, is unstable and not present in nature. REEs in nature are usually present in a 3⁺ valence state. Cerium (Ce) and Europium (Eu) are also present in oxidation states of 4⁺ and 2⁺, respectively. REEs can be divided into two groups: light and heavy REEs. The distinction is based on their separability (physical/chemical) and ion radius. The light REEs are La, Ce, Pr, Nd, Sm, Eu and Gd, whereas the heavy REEs are Y, Tb, Dy, Ho, Er, Tm, Yb, Lu and Sc.

1 Application of REEs in the Agriculture of China

China has the largest reserves of REEs in the world (80%) and is a major producer of REEs for the world market. Since 1990, REE fertilizer has been widely used in more than 20 Chinese

provinces (Yu and Chen 1995). REE fertilizer intended for agricultural production was predicted to cover over 16-20 × 10⁶ ha of agricultural land in 1995 in China (Wu and Guo 1995).

1.1 Types of Chinese fertilizers with REEs

There are basically three kinds of Chinese fertilizers, each of which contains different REEs. They are respectively: Changle-Yizhisu (CY), which contains nitrate forms of REEs (Table 1); Nongle (NL), which contains chloride forms of REEs chloride and its main component belongs to REEs (38% as RE₂O₃); and MAR (rare-earth complex of mixed amino-acids), which contains 17 amino-acids together with elements of La, Ce, Pr and Nd.

Table 1: The single rare-earth elements content in CY (%)

La ₂ O ₃	CeO ₂	Pr ₆ O ₁₁	Nd ₂ O ₃	Sm ₂ O ₃	Eu ₂ O ₃	Gd ₂ O ₃	Σ RE ₂ O ₃
19.78	4.66	1.86	5.40	0.34	0.07	0.08	32.19

(He et al 1998)

1.2 The effects of REEs on yield and quality of crops

Hong et al. (1996) reported that the REEs can raise the output of wheat 4~10% every year in a continuous ten-year period after applications of REEs of 600 g/ha-yr. Other studies also showed that plant growth could be increased by REEs at appropriate concentrations. For example, one report showed that the seed germinating rate of winter wheat was enhanced 8~19% after blending with 30-50 mg/L REEs (Wu et al. 1984). Another study demonstrated that the germinating rate of scallion and onion was increased 13~14%, and that of eggplants was 23~45% higher after blending with 50~500 mg/L REEs (Zheng et al. 1993). REEs also increased the seedling and root growth. Ning and Xiao (1989) reported that REEs (<5 mg/L, R₂O₃) could enhance the new root growth of rice, but it would inhibit rice from growing when REEs content is more than 5 mg/L. Table 2 shows the effects of REEs on the other main crops.

The effects of different REEs on plant growth are different. Tang and Tong (1988) showed that the radical growth in Chinese cabbage could be increased by low-concentration La, Ce, Pr and Nd (0.05~1 mg/L), especially by Ce, but that high concentration of REEs (>10 mg/L) prevented root growth.

* Reprint from ESPR – Environ Sci & Pollut Res (DOI: <http://dx.doi.org/10.1065/espr2001.05.065>)

Table 2: The effects of REEs on crops

Crops	Effects on yield			Effects on quality
	Range %	Average %	kg/ha	
Maize	6~12	8	540	Improved by 3 g per 1000 grains
Potato	10~14	13	2850	Improved starch by ~1%
Rape	14~24	15	165	Improve oil content by ~1%
Ramine	7~15	10	82.5	Improve fiber content by 10~12%
Flax	flax: 8~10 seeds: 10~14	10 12	360~450	Improve fiber intensity by 10%
Reed	11~19	13	2550	Improve fiber content by 1.8~4.6%
Chinese gooseberry	12~25	17	4~8 kg (per plant)	Improve sugar content by 1.3~2.9%
Haw		25	3 kg (per plant)	Improve anthocyanin content by 16%
Banana	8~14	10	2700	Improve sugar content by 3~4%; Vc 5%
Astragali	12~19	15	5250	Improve sugar content by 3~4%; Vc 5%
Alfalfa		17	750~1950 (dry weight)	Improve 1000-grain weight by ~5%
Mushroom	10~13	11	6 kg/m	Improve coarse protein by 3~9%

(Guo 1993)

Table 3: The application methods and concentrations of REEs

Crops	Application methods and amounts
Wheat	Spray: 600 mg/L (end of March until 10 April)
Maize	Blending seeds: 3 g/kg Immerse seeds: 8 g/kg
Potato	Blending seeds: 6 g/kg
Rape	Blending seeds: 5 g/kg
Ramine	Spray : 100~300 mg/L (seedling period)
Flax	blending seeds: 600 g/ha spray: appear bud period
Reed	spray:600~900 g/ha (seedling or flower period)
Chinese gooseberry	spray: 700 mg/L (flower and young fruit period)
Haw	spray: 400 mg/L (flower period)
Banana	spray:300~500 mg/L (seedling and young fruit period)
Astragali	spray: 300 mg/L (seedling period)
Alfalfa	blending seeds:100~300 mg/kg
Mushroom	spray: 50 mg/L

(Guo 1993)

Table 4: Accumulation of REEs in spring wheat of mature age (mg/kg)

	Root	Stem	Leaf
Check	37.9	0.42	3.44
Treated	50.1	0.55	5.73

1.3 REE application methods and the concentration to be used

REEs have been used as fertilizers by blending seeds, immersing seeds and spraying foliar. Different crops used different amounts of REEs and different methods of treatments. REEs must be used as fertilizer every year, otherwise it would have no effect. Table 3 shows the application methods and the amount of REEs.

1.4 Contents, distribution and accumulation of REEs in crops

Sun et al. (1986) measured the content of REEs in the main crops from 8 Chinese provinces. He found that the amount of REEs are 0.39 ± 0.1 mg/kg in rice, 0.13 ± 0.1 mg/kg in corn 1.15 ± 0.18 mg/kg in wheat, 0.5 ± 0.12 mg/kg in vegetable and fruits, and 12~1.58 mg/kg in beans.

The accumulation of REEs is highest in roots (88~90%), less in the crust and stem (10~12%), and lowest in leaves (Hong et al. 1996). After the application of REE fertilizer for 11 yrs, the REE content in the root, stem and leaves of spring wheat have been enhanced, but there are no clear changes in the seed compared with check-ups (Table 4 and 5, Liu et

Table 5: REE content in grains of spring wheat (mg/kg)

	La	Ce	Nd	Sm	Eu	Tb	Yb	Lu
Check	0.005	<0.0075	<0.004	0.0007	0.0006	<0.0002	<0.0008	<0.0002
Treated	0.004	<0.0075	<0.004	0.0007	0.0004	<0.0002	<0.0008	<0.0002

al. 1996). This result was in disagreement with those from Hong et al. (1996) who reported that there was no obvious accumulation in the stem of spring wheat after the application of 600 g REEs / ha-yr for 10 yrs.

2 Physiological Effects of REEs on Plants

2.1 Effects on nutrient metabolism

Although there is no clear evidence to show that REEs are necessary for plants to grow, many studies suggest that REEs can stimulate plants to absorb, transfer and assimilate nutrients. Ning and Xiao (1989) reported that after using REEs as fertilizers, the absorption of rice for N, P, and K is increased by 16.4%, 12%, and 8.5%, respectively; The absorption of sulfate by soybeans is also augmented after the application of REEs. Lai (1989) found that tomatoes absorb 8.13% more NO_3^- after blending seeds in 50 mg/L REEs by the ^{15}N trace technique. Tang (1998) reported that P and K contents are enhanced 10.34% and 15.42% after spraying tomato seedlings by 5 mg/L CeCl_3 . These results suggest that the effects of REEs on improving the absorption of nutrient elements depend upon the methods used in treating the plants. Spraying REEs on plants is commonly thought to be a better method than blending seeds in REEs (Wang 1994).

The metabolism of nutrients in plants has been shown to be significantly increased by REEs. Nitratase activity in peanuts and tomatoes was enhanced markedly by spraying REEs (Guo 1988). The transfer rate for N from the inorganic to the organic form was accelerated, which is a benefit for protein synthesis and the regulation of nutrient balance. In addition, Yang and Zhang (1986) showed that nitratase activity in the leaves of winter wheat was enhanced by 37~75% after blending seeds in REEs, and the yield was improved by 15.52% compared with control groups. After the application of REEs, the number of root nodules was increased significantly, and the activity of nitrogen-fixation was improved by 24%. As a result, the absorption of N by legumes was enhanced significantly by REEs (Wu et al. 1985).

However, it was noted that solutions containing greater than 10 μm La, Ce and Yb concentrations severely inhibit plant growth (Diatloff et al. 1995a 1995b 1995c, Ishikawa et al. 1996, Kinraide et al. 1992). REEs have been found to disturb the metabolism of calcium and, to a lesser extent, magnesium (Nair et al. 1989). They have also been reported to replace and compete with Ca for binding to proteins, and affect the stability of cell membranes (Hu and Ye 1996). The interference of calcium function by REEs, especially lanthanum, is probably an important cause for their toxicity. In addition, Velasco et al. (1979) suggested that REEs replaced the essential element boron from its active sites and induced boron deficiency. Diatloff et al. (1995c) demonstrated manganese deficiency in mungbean plants exposed to solutions containing $>0.63 \mu\text{m}$ Ce. The high-concentration REEs could destroy cell membrane stability and increase cell permeability, which may lead to a K^+ flux and deregulation of nutrient metabolism (Chang 1991).

2.2 Effects of REEs on photosynthesis

REEs have been found to markedly influence photosynthesis. La, Ce, and Pr of less than 50 mg/L could increase photosynthesis in nitrogen-fixation alga. However, if their concentration is more than 50 mg/L, they inhibit photosynthesis (Wang et al. 1985). Blending seeds with REEs increased the chlorophyll content and rate of photosynthesis in sugar beets by 4.7% and 31.8% (Xie and Chen 1984). Spraying 200~800 mg/L of REEs Spraying on pepper foliars improved the total chlorophyll content of chl_a, chl_b (He et al. 1998). The optimum concentration of REEs was found to be around 400 mg/L, and REEs could be harmful at very high concentrations. Chu's (1996) experiments showed that the optimum concentration of CeCl_3 on the photosynthesis for wheat seedling was 0.2 to 0.5 mg/L, while 10 mg/L became harmful (nutrient solution culture). For cucumbers, the optimum concentration was in the range of 1 to 5 mg/kg, and for sunflowers 15 mg/L. REEs could also increase the assimilation transporting to seeds (Liu and Wan 1993).

The principle of REE effects on photosynthesis was not very clear. Chu et al. (1996) reported that CeCl_3 could accelerate the synthesis of chlorophyll a (Chl_a) and protein in *spirulina platensis*, and enhance the activity of oxygen evolution.

2.3 Effects on resisting stress

Greater biomass and increased root growth were observed after the exposure to low lanthanide concentrations (Velasco et al. 1979, Diatloff et al. 1995c, Pham Thi Huynh et al. 1997), especially under stress (Guo et al. 1988). REEs could reduce the amount of electrolyte effusion and enhance the concentration of proline in leaves and seedlings of wheat under -8°C for one hour, suggesting that REEs could enhance the ability of resisting cold by wheat seedlings. Some researchers reported that REEs could increase the ability of resistance of drought by maize (Wen et al. 1992), resistance of acid rain by spinach (Yan et al. 1998, 1999), and resistance of metal stress (Zhou et al. 1998, 1999).

Through our experiments and those of others (Yan et al. 1999, Zhou et al. 1998), it was found that REEs could enhance the antioxidant potential of plants. Wang et al. (1997) indicated that Ce^{3+} could reduce O_2^- to H_2O_2 , and could itself be oxidated to Ce^{4+} . Ce^{4+} could oxidate O_2^- to O_2 , while it itself reduces to Ce^{3+} . These results may be the mechanisms of REEs to resist stress.

3 Environmental Behaviors of REEs in Soil

An investigation of the effects of REEs on environments has been limited due to the little historic use and the lack of sensitive analytical techniques. Much of the available literature concerns the distribution and abundance of REEs from the geochemical and mineralogical point of view. In the past 20 years, REEs turned out to be promising elements due to their excellent properties for fine chemistry in modern industry. Therefore, environmental contamination from the wide-spread use of REEs is likely to increase. In addition, the intensive application of REEs in agriculture in the 80's in China requires a thorough investigation on their chemical behavior in the soil.

The environmental behavior of REEs in soil is dominated by their low solubility (Weltje 1997). Fluorides, carbonates, phosphates and hydroxides may form complexes with neutral REEs with low solubility, resulting in low dissolved concentrations in the aqueous phase of ecosystems. In solution, REEs may be complexed with inorganic ligands (e.g. carbonate, sulfate), organic ligands (e.g. humic and fulvic acids) and, at a high pH, with hydroxyl ions.

3.1 Adsorption and desorption

The adsorption and desorption of REEs in soil and in synthetic oxides has been studied (Ran and Liu 1992, 1993). The isothermal adsorption of REEs on major types of soil in China, on the synthetic metal oxides, as well as on kaolinite could be simulated by Freundlich, Temkin and Langmuir adsorption equations. The maximum capacity of REEs adsorption (Q_m) were 57.0, 12.7, 7.90, 7.65, 5.13, 1.96, 1.62, and 0.90 mg/g for δ -MnO₂, black soil, chernozem, amorphous iron oxide, yellow brown soil, red soil, laterite, and kaolinite, respectively. Table 6 shows some of the results.

The adsorption capacity of REEs depends on the clay type and the count of amorphous iron and manganese oxides, the latter having the high adsorption ability. In contrast, the desorption of REEs is generally very low, with the exception of REEs being adsorbed by red soil and yellow brown soil (Peng and Wang 1996).

The influence of acid precipitation on the environmental chemical behavior of REEs was studied recently by a simulation experiment (Chen et al. 1995). Acidity of the solution and volume of acid apparently caused the leaching of REEs from soil. The leaching property varies with type of soil and individual rare-earth element. For examples, La, Gd and Y have found to behave differently in the leaching process.

3.2 Water-soluble rare-earth elements (WSREEs) in soil

WSREEs in soil are the most active forms of REEs and play the most important role in both the environmental behavior and biological effect. Thus, studying the content, distribution and the ratio to total rare-earth elements is most important.

Content of WSREEs in soil: Most of the compounds bearing REEs in soil, as mentioned above, are insoluble substances. The order of solubility ranged from 10^{-7} to 10^{-4} mol/L. Because of adsorption, coordination, and co-precipitation in soil-water systems, the concentration of REEs in soil solu-

tion was usually lower than 1 mg/kg. The content of WSREEs in soil was lower than 100 mg/L IN most cases, and varied widely both in different soils and different layers of the same profile. The average of WSREEs content in soil was 10–20 mg/kg.

Distribution of WSREEs in profiles: An interesting phenomenon was found that decreased the content of REEs from the top to the bottom layer for red soil, but increased it for brown soil and black soil (Zhu et al. 1996). Although the extract reasons for this phenomenon are unclear, the difference in rainfall, contents of organic matter and clay, and pH might be the main factors which cause the differences of the WSREE distribution patterns. Rainfall in southern China is much higher than that in northern China. But in northern China, WSREEs of the higher layers were leached into lower layers, where they were adsorbed. The higher contents of organic matter and clay could hold more ionic REEs in soil. However, the exact mechanism for causing this difference needs to be studied further.

The ratio of WSREEs and REEs of soil: Generally the fraction of WSREEs is about 10% or lower than the total REEs in soil samples (Zhu et al. 1996).

3.3 Chemical forms of extraneous REEs in soil

The forms of REEs present in various types of soil have been evaluated by Tessier-sequence extracted techniques (Zhu and Xi 1992). The results showed that the percentile distribution of association forms of REEs in soil correlates closely to the physical-chemical properties of soil itself. Among these forms, the exchangeable fraction (CREE) ranges from a trace to 24.1 mg/kg and amounts to about 0–10.5%. Amorphous Fe-Mn oxides associate with about 30% to the total REEs in soil, ranking the highest. This form could accumulate in the lower layer of soil. For all of the rare-earth elements, the residual form accounts for the majority, while the water-soluble and exchangeable fractions are very small. In soil, average percentages of the residual fraction vary from 45.2% to 94.7% for individual RFE. This shows that REEs are mainly bound in mineral lattices in the soil. The behaviors of REEs during weathering and soil-forming process show that they are removed from primary minerals. The soil matter has inherited REE characteristics from the source of parent material (rocks). The residual form is stable under natural conditions, while the other forms are unstable (easily activated). The non-residual form of REE is mainly bound to carbonates, Fe-Mn

Table 6: Desorption of absorbed REEs by soil and minerals

Samples	Adsorption(A) $\mu\text{g/g}$	Desorption(B) $\mu\text{g/g}$	Net (A-B)	Ratio of B/A
Amorphous Fe oxides	6275-6625	Trace-4.25	6275-6621	1
δ -MnO ₂	5250-31500	Trace-trace	5250-31500	0
kaolinite	344-850	222-740	106-122	64-87
laterite	422-1525	81-798	341-727	19-52
red soil	731	650	81	89
yellow brown soil	1030	900	121	90
black soil	1043	10	1033	1
chernozem	1041-6238	11-2078	1030-4160	1-33

oxides and organic matter. In different soil, these forms account for 0.75-8.04%, 0.69-28.0% and 1.84-26.5% of the individual REE content, respectively. The amount of the seven forms for all of the REEs demonstrate the following relationship: residual>>bound to organic matter>bound to Fe-Mn oxides>bound to carbonates>>exchangeable and water soluble (Zhang et al. 1996).

Liu et al. (1999) studied the transformation of rare-earth elements in soil. The results showed that high-concentration (2-60 mg/L) REEs added to soils was rapidly converted to other forms. As time went on, soluble exchangeable rare-earth elements decreased rapidly. Organic complex REEs remained unchanged at first, and then increased. Fe/Mn oxide-bonded REEs initially increased, but subsequently decreased. The residual fraction was stable.

4 Prospects

Russian scientists have worked on the pollution of REEs in the soil from phosphorus fertilizer production. An increased storage in agricultural plants was found (Volokh et al. 1990). The content of REEs in the hair of workers and local residents was found to be higher than in the neighboring ones. Higher REE contents in human hair reflected well the REEs transport from soils and plants to human beings (Markert 1987, Ichihashi et al. 1992). These observations are indicative for evaluating the possible contamination of the environment by REEs resulting from an increased usage in agriculture.

To fully understand the effects of REEs in agricultural application, environment and human health, we suggest that future research in several areas is needed. First, the dynamic changes and fates of REEs used in fertilizers should be thoroughly investigated. Second, the indirect effects of REEs on human health and ecosystems through their use in agriculture should be explored. Third, it is necessary to make an assessment of REEs as hazards in their use in agriculture and to develop standards for soil quality.

Acknowledgements. The work was supported by a grant from the Chinese National Science Foundation (29890280-1). We thank Dr. Wang for improving the English.

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Received: November 30th, 2000

Accepted: March 3rd, 2001

Online-First: May 11th, 2001

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