磷石膏の土壌反応と作物生育への影響

著者: 髙須 栄一
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氏名（本籍） 髙 須 栄 一
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論文審査委員 （主 塚） 教 授 三枝 正 彦
（副 塚） 教 授 國 分 牧 衛
助教授 南 條 正 巳

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Chapter 1 Introduction

Phosphogypsum (PG) is obtained as a by-product when phosphoric acid is produced from rock phosphate, and it primarily comprises hydrated gypsum (CaSO₄·2H₂O). During the process of wet phosphoric acid production, several times of PG is produced along with the end product; therefore, a large deposit of PG still remains in factories. PG comprises only around 4% of the total agricultural use in a few countries or areas, while its world production is estimated to be approximately 125 million Mg/year (Rechigl 1995). Therefore, the agricultural use of PG is very important from the viewpoint of the stable reuse of industrial by-products.

The agricultural use of gypsum has been primarily examined to ascertain the improvement in the chemical and physical properties of deteriorated soils. PG has been used as an effective ameliorant for subsoil acidity in acidic and low base status soils, i.e., Oxisols and Ultisols (Shainberg I et al. 1989). Moreover, it is well known that gypsum application suppresses soil dispersion in sodic soils (Harrison WJ et al. 1992). Since Saigusa et al. (1991) reported that PG is an effective ameliorant for subsoil acidity in non-allophanic Andosols, many experimental studies on the agricultural use of PG have been conducted in Japan, particularly for improving sulfur nutrition in Orchardgrass, controlling the decomposition of rice straw manure, and improving the calcium (Ca) nutrition and constraint effect of the russet scab in potato (Toma M et al. 1995, 1997, and Toma M and Saigusa M 1997). However, in Japan, most studies on the effect of PG application have been conducted using nonallophanic Andosols, a few studies have been conducted on alluvial soils and terrace soils. Therefore, it is necessary to examine the effect of PG in other soil types, particularly in allophonic Andosols and other soils distributed in alluvial areas such as Inceptisols and Entisols.

Leaching of the base such as Ca and the resulting soil acidity are problems that occur in Andosols. Therefore, in Japan, liming is performed by using calcium carbonate or dolomite. However, they do not supply a sufficient amount of Ca to crops due to its slight solubility. Since PG is moderately soluble in water (0.2 g L⁻¹) in comparison with calcium carbonate or calcium chloride, it is a promising Ca source for crops that require a large amount of Ca. Although many attempts have been made to determine the amount of PG required supplying Ca to crops, few studies have been conducted with a focus on growth response such as root development. In addition to the supply of Ca, it is expected that PG application can improve the physical properties of the soils for upland crops converted from paddy fields. In addition, it is also necessary to clarify the effect of PG in a paddy field mainly distributed in an alluvial area because the soils converted from paddy fields rotationally again reconvert to paddy fields.

Therefore, in order to clarify the effect of PG application on Andosols and other soils distributed in an alluvial area, which are widely distributed in Japan, the following examinations were conducted by considering the relationship between the changes in both the chemical and physical properties of the soils and crop growth with special reference to their root growth.
Chapter 2 Effect of phosphogypsum application on the chemical properties of alluvial soils and terrace soils

It is necessary to examine the basic reaction of PG in the soil, particularly in the alluvial and terrace soils such as Inceptisols and Entisols in Japan because the information on these soils is limited. Therefore, the changes in soil pH after PG application were examined with special reference to the acidic character of the soil. Ten cultivated soil samples, which were classified into alluvial and terrace soils, were used in this experiment. PG was applied at the rates of 0, 1.0, 2.0, 4.0, and 8.0 g kg⁻¹ air dry soil.

Although the soil pH decreased in proportion to the application rate of PG regardless of the soil types (Fig. 1). There was a significant negative correlation between the apparent decrease in the soil pH and the amount of constant charge in the soil at the 5% level (Fig. 2). In addition, the increase of the Al³⁺ concentration in the soil solutions with PG application was less than 9% of the decrease of both exchangeable Al and y₁ in all the soils tested. Therefore, it indicates that the Al released could be irreversibly changed to other forms such as polymerized Al, as previously reported on non-allophanic Andosols.

Chapter 3 Reaction of phosphogypsum in Andisols and its effect on crop growth

Section 3-1 Evaluation of plant-available calcium

Exchangeable Ca does not necessarily correlate with Ca uptake by crops. Therefore, in order to evaluate Ca availability to plants, the effect of PG application on the chemical properties of soil and Ca uptake by Komatsuna (Brassica rapa L. cv. Natsumakuten) was examined in comparison with that of calcium carbonate with special reference to both exchangeable and water-soluble Ca in the soil.

Exchangeable Ca increased in a similar manner in both the calcium carbonate and PG plots. On the other hand, water-soluble Ca increased in proportion to the PG application rate (Fig.3), and the maximum increase was approximately 5 times higher than that observed in calcium carbonate plot.

In Komatsuna plant, PG application affected the root growth much stronger than the top growth; the fresh weight of roots grown in PG plots was 6%–31% greater than that of roots grown in calcium carbonate when the same amount of CaO was applied. The Ca content of Komatsuna plants grown in PG plots was greater than that in calcium carbonate plots. Adding to this, Ca uptake by Komatsuna plant was closely correlated to the water-soluble Ca content (r² = 0.8461) of the soil in comparison with the extractable Ca content (r² = 0.4832) of the soil (Fig.4); Therefore, it was clarified that water-soluble Ca was more suitable as an index for plant-available Ca than exchangeable Ca in soil.
Section 3-2 Reaction of PG in Andisols with different colloidal components

In Section 3-1, it was clarified that PG application to allophanic Andosols was an effective method for supplying Ca to crops and the evaluation of plant-available Ca became possible by measuring the water-soluble Ca in the soil. However, it was also predicted that the effectiveness of PG application on supplying Ca would be affected by the colloidal components in Andosols. Therefore, in this section, the effect of Ca supply in Andosols with different colloidal components was examined with special reference to water-soluble Ca. Five virgin Andosols with different inorganic and organic colloidal components were used in this experiment. PG was applied to the soils at the rates of 0, 0.3, 0.6, and 1.2 CaO g kg\(^{-1}\).

In all the soil samples, the water-soluble Ca content increased with an increase in the PG application rate; however, the increment differed among the soil types (Fig.5). The increment in the water-soluble Ca content of non-allophanic Andosols was relatively higher than that of allophanic Andosols. The apparent increment in the water-soluble Ca content tended to decrease exponentially with an increase in the allophane content of the soil (Fig.6). Furthermore, on comparing between non-allophanic Andosols with less than 50 g kg\(^{-1}\) of allophane content, it was deduced that the apparent increment in the water-soluble Ca content of humic non-allophanic Andosols was lower than that of non-humic soils by 0.1 cmol\(_c\) kg\(^{-1}\).

Section 3-3 Effects of PG application to nursery media prepared from Andisols on growth and calcium uptake of melon seedlings

Based on the results of Section 3-1, it was clarified that PG application could supply Ca to crops without causing salt injury and could also promote the root growth of crops. It is well known that the melon plant \((Cucumis melo \ L.)\) is a crop with a high Ca requirement and that root development in this plant is a key to promote initial growth. Therefore, considering the above-mentioned advantageous PG effects, the effect of PG application to nursery media prepared from Andisols on the quality of melon seedlings was examined in this section. Three melon varieties were used in this experiment and PG was applied to the nursery media at the rates of 0, 2.0, 4.0, and 8.0 g L\(^{-1}\).

PG application increased water-soluble Ca in the nursery media from 1.7 to 5.2 cmol\(_c\) L\(^{-1}\). Both top growth and root growth of melon seedlings were enhanced by PG application regardless of the melon variety (Table1). Although Ca uptake by melon seedlings was promoted by PG application in all the varieties, the relationship between Ca uptake and growth response of roots was different among varieties (Fig.7). Considering the root growth, it was decided that the optimum PG application rate for melon seedlings is 4 g L\(^{-1}\).
Section 3-4 Relationship between the nutrient uptake in cucumber and the root response to phosphogypsum application

In the results stated in Sections 3-1 and 3-2, the effect of PG application on both Ca supply and root growth of crops was clarified. In this section, the effect of PG on the growth of cucumber was examined with a special reference to the soil-plant interaction.

The pot experiment with allophanic Andosols was conducted using the cucumber variety, *Cucumis sativus* L. cv. Sharp-1. Calcium compounds such as calcium carbonate, calcium chloride, and PG were used, and their application rates were 0, 0.15, 0.3, 0.6, 1.2, and 2.4 CaO g kg⁻¹.

The dry weight of the root increased on the application of both calcium chloride and PG compared with that of the control, regardless of the application rate (Table2). The amount of rhizosphere soil differed among experimental plots and increased proportionally with the PG application rate, reaching a value approximately 4 times greater than that of the control. There was a positive correlation between the amount of rhizosphere soil retained by the roots and Ca/Total cation ratio of the soil solutions at 0.01% level (Fig.8). Furthermore, microscopic observation showed that root hair was well developed in the plants with PG application compared with the root hair in the control plot. From the result of the examination on the relationship between root-related factors and the nutrient uptake of *Cucumis sativus* L., the best correlations was obtained between the net amount of rhizosphere soil retained by the cucumber root and the nutrients uptake, such as N, P₂O₅, Ca, and S.
Chapter 4  Reaction of phosphogypsum in paddy soil and its effect on crop growth

Section 4-1 Effect of phosphogypsum application on the chemical and physical properties of the soils converted from paddy field and on the growth of soybean

In Chapter 3, the effectiveness of PG on Ca supply in Andosols was clarified from the viewpoint of root growth. On the other hand, it is very important to improve field permeability in addition to Ca supply in the soils converted from paddy field, in where poor drainage becomes a limiting factor for a stable soybean production. Therefore, in order to clarify the effect of PG application in the soils converted from paddy field, the following experiments were carried out.

A. Effect of phosphogypsum application on the chemical and physical properties of the soils converted from paddy fields and on the root growth of soybean

The experiment was conducted using Glycine max L. Miyagishirome in the soils converted from a paddy field in Furukawa, Miyagi prefecture. Calcium carbonate, powdery phosphogypsum (PPG), and granulated phosphogypsum (GPG) were applied at the rate of 0.3 Mg CaO ha$^{-1}$.

Both PPG and GPG application remarkably increased the amount of water-soluble Ca in soil compared with the control. On the other hand, the measurement of the three soil phases in the PPG plot showed a 4.2% increase in the gaseous phase and a 3.5% decrease in the solid phase as compared to the control. The soil dispersion ratio had decreased significantly with the application of any kinds of Ca-containing compound. The infiltration rate in the field was measured, and the following order of decreasing water permeability was obtained based on the n-value in infiltration curves: PPG > GPG > calcium carbonate > control (fig.9). Furthermore, the remarkable decreasing of the stagnant water in the furrow was observed in PPG plot after the heavy rainfall (Fig.10).

Top growth of soybean such as the length of the main stem and its node number were significantly increased with PPG application (Table3). In addition, the descending order of the total root length corresponded with the descending order of the field permeability. The amount of Ca uptake by soybean plant was increased in the PPG and GPG as compared to the control. The results of root distribution at the harvest time revealed that root development was vertically and laterally promoted in the PPG plot compared with that in the control (Fig.11).

B. Evaluation of the effectiveness of phosphogypsum in different soil types

In the previous section, it was clarified that the effect of PG in the soils converted from a paddy field. However, it was predicted that these effects would differ among the soil types. Therefore, in this section, the effect of PG on soil dispersion in different soil types was examined by a simplified method that measures the filtration speed in soil suspensions. PG was applied at rates of 0, 0.5, 1.0, and 2.0 g kg$^{-1}$. Effect of PG on soil dispersion considerably differed among the soil samples tested. The suppression of soil dispersion by PG was greater in non-Andosols. It was also remarkably higher, particularly in Takadate soil with dominant 2:1 type clay minerals.
Section 4-2 Effect of phosphogypsum application on the chemical properties of paddy soil and the growth of rice plants

In section 4-1, it was clarified that the effect of PG in the soils converted from a paddy field. However, it is also necessary to examine the effect of PG in paddy fields. And the injury induced by the generation of hydrogen sulfide following PG application has been considered. Therefore, in this section, both positive and negative effects of PG on rice growth were examined, particularly with respect to soil solution components and the root growth of rice.

A. Effect of phosphogypsum application on the chemical properties of flooded soil and on the initial growth of rice plants

The effect of PG on the initial growth of rice plants was examined using five soils with different colloidal components. Pot experiments were carried out on Oryza sativa L. Koshihikari by using 1/5000 a Wagner pots, and the application rates of PG were 0, 1.0, and 2.0 g kg⁻¹.

NH₄⁺ concentration in the soil solution had been increased with PG application compared with that in the control, irrespective of the soil sample used. Furthermore, the number of tiller in rice plants tended to increase with PG application. The extent of increase in the number of tiller corresponded well with the increase of NH₄⁺ in the soil solutions.

B. Effect of PG application on the growth of rice plant and its productivity

In this subsection, the effect of PG on rice productivity and the hydrogen sulfide injury due to the PG application in a paddy field were examined.

The experiment was conducted in a paddy field (Typic Melanaquands) by using Oryza sativa L. Hitomebore. The fields where PG was applied at a rate of 0 (PG0), 1.0 (PG100), 2.0 (PG200), and 5.0 (PG500) Mg ha⁻¹ were used as the main plots, while those with nitrogen topdressing were considered as subplots.

Similar to the pot experiments, the number of tillers with PG application increased as compared with that in the control, particularly in the case of PG500 (Fig.12). The rice roots were classified according to their root color prior to heading, and the ratio of the black roots apparently increased in proportion to the amount of PG applied (Fig.13). However, following the staining of a cross section of a black root by Evans blue, cell death and morphological changes in the stele were not observed microscopically. Therefore, it was considered that the proportion of the black root was not well correlated with rice yield, and PG application does not necessarily induce hydrogen sulfide injury. The ear number and total number of unhulled rice increased in proportion to the amount of PG applied, resulting in an 8% increase in brown rice yield in PG500 (Fig.15). Additional effects of nitrogen topdressing on the brown rice yield were also observed except PG500 plot.
5. General conclusion

The reaction of phosphogypsum (PG) in soil and its effect on crop growth in Andosols and other soils distributed in an alluvial area were examined with special reference to root growth. The results obtained in this study are as follows:

1. By examining the reaction of PG in alluvial soils, it was clarified that the soil pH decreased due to PG application and the extent of this decrease was reflected by the amount of constant charge in the soils.

2. By examining the effect of PG on Ca supply in Andosols, the following aspects were clarified:
   (1) The amount of water-soluble Ca in the soil can be practically determined and is useful as an index of plant-available Ca.
   (2) The apparent increase in the water-soluble Ca content with PG application was influenced to a greater extent by the allophane content than the humus content.
   (3) The application of PG to nursery media prepared from Andosols is a promising method for supplying Ca to melon seedlings and consequently promoting root development, and the optimum application rate is 4 g L⁻¹.
   (4) PG application contributed to the nutrient uptake of cucumber by improving root hair development, which resulted in an increased amount of retained rhizosphere soil.

3. By examining the effect of PG in both the soybean soils converted from a paddy field and the paddy field soil, the following aspects were clarified:
   (1) Powdery phosphogypsum (PPG) application was demonstrated to be a practical technique for improving field permeability. Further, its promotional effect on root growth continued from the initial growth stage to the harvest time. Moreover, it was confirmed that the effect of PG on soil dispersion was the most pronounced in clayey soil with dominant 2:1 type clay minerals.
   (2) PG application promotes the initial growth of the rice plant and shows the potential to increase rice productivity with and without additional nitrogen topdressing.

From the abovementioned, it was clarified that PG application is significantly effective on supplying Ca to crops in Andosols, particularly in allophanic Andosols, resulting in a considerable improvement in root growth. Furthermore, the effectiveness of PG application as both a Ca source and an ameliorant of soil physiology in the soils converted from paddy fields as well as the possibility of the use of PG in paddy fields were clarified by considering its positive and negative effects on rice growth.
Fig. 1 Changes in pH of different soil types with PG application

Fig. 2 Relationship between the constant charge of soil and soil pH changes

Fig. 3 Changes in the exchangeable and water-soluble Ca in soil with application of Ca materials

Fig. 4 Relationship between the water-soluble Ca in soils and Ca uptake by *Brassica rapa* L.

Fig. 5 Effect of PG application on the water-soluble Ca in Andisols

Fig. 6 Relationship between the allophane content and apparent increase amount in water-soluble Ca (ΔCa) in Andisols treated with PG
Table 1  Effect of PG application on the growth of melon seedlings at 24 days after sowing.

<table>
<thead>
<tr>
<th>Variables</th>
<th>Treatment</th>
<th>Leaf number</th>
<th>Plant height (cm)</th>
<th>Dry weight (g plant⁻¹)</th>
<th>Dry weight (g plant⁻¹)</th>
<th>TR ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>Top</td>
<td>Root</td>
<td>Top</td>
<td>Root</td>
</tr>
<tr>
<td>Amus</td>
<td>PG0</td>
<td>3.4 a</td>
<td>17.0 a</td>
<td>1.84 a</td>
<td>0.40 a</td>
<td>4.8</td>
</tr>
<tr>
<td></td>
<td>PG2</td>
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<td>0.36 a</td>
<td>4.7</td>
</tr>
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<td></td>
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<td>22.1 a</td>
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<td>0.54 a</td>
<td>4.0</td>
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<td></td>
<td>PG8</td>
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<td>2.02 a</td>
<td>0.43 a</td>
<td>4.8</td>
</tr>
<tr>
<td>Earl's</td>
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<td>1.92 a</td>
<td>0.30 a</td>
<td>6.4</td>
</tr>
<tr>
<td></td>
<td>PG2</td>
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<td>12.4 a</td>
<td>2.05 a</td>
<td>0.32 a</td>
<td>6.5</td>
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<tr>
<td></td>
<td>PG4</td>
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<td>2.23 a</td>
<td>0.44 a</td>
<td>5.1</td>
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<td>Midorishima</td>
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<td>0.44 a</td>
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</table>

*Means with the same letter are not significantly different at the 5% level by Student's t-test.

Table 2  Effect of different Ca salts application on cucumber growth.

<table>
<thead>
<tr>
<th>Experimental soils</th>
<th>Leaf number</th>
<th>Plant height (cm)</th>
<th>Dry weight (g plant⁻¹)</th>
<th>Top</th>
<th>Root</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ca source</td>
<td>Application rate (CaO g Mg⁻¹)</td>
<td>Top</td>
<td>Root</td>
<td>Top</td>
<td>Root</td>
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<td>Control</td>
<td>0.0 a</td>
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<td>Calcium carbonate</td>
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<td>2.92 a</td>
<td>6.21 a</td>
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<td>Phosphogypsum</td>
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<tr>
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<td>3.03 a</td>
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<td>65.4 a</td>
<td>2.91 a</td>
<td>6.15 a</td>
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</tbody>
</table>

Fig. 7  Relationship between Ca uptake and root growth of melon seedlings.

The relative root growth is expressed as the increment against control in dry matter weight of root.

Fig. 8  Relationship between the Ca/TC ratio in the soil solution and the amount of rhizosphere soil retained by the roots of Cucumis sativus L.

Fig. 9  Infiltration curve.

*Measurement under the condition of 0.49 m³ m⁻³ in volumetric water content.
**Data shows the mean of 5 replications.
Fig. 10 Comparison of stagnated water at 2 hr. after heavy rainfall (35mm precipitation)

Table 3 Effect of phosphogypsum application on the growth of soybean

<table>
<thead>
<tr>
<th>Treatment</th>
<th>Application rate (CaO Mg ha⁻¹)</th>
<th>Length of the main stem (cm)</th>
<th>Node number of the main stem (number plant⁻¹)</th>
<th>Total root length (m plant⁻¹)</th>
<th>Shoot Dry Weight (g plant⁻¹)</th>
<th>Root Dry Weight (g plant⁻¹)</th>
<th>Nodule</th>
</tr>
</thead>
<tbody>
<tr>
<td>Control</td>
<td>0</td>
<td>12.7a 34.1a 94.4a 2.4 a 7.7 a 9.79 a 0.60 a 0.0057 a 59.7 a 4.66 a 0.158 a</td>
<td></td>
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<td></td>
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<td></td>
</tr>
<tr>
<td>Calcium carbonate</td>
<td>0.3</td>
<td>13.3 a 36.0 a 96.6 a 2.4 a 7.9 a 17.0 a 11.1 ab 0.60 a 0.111 a 54.4 a 4.43 a 0.155 a</td>
<td></td>
<td></td>
<td></td>
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</tr>
<tr>
<td>PPG</td>
<td>0.3</td>
<td>12.8 a 35.0 a 102.3 b 2.4 a 7.5 a 17.6 b 13.1 b 0.64 a 0.122 a 73.3 a 6.50 a 0.197 a</td>
<td></td>
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Data are expressed as the mean ± standard deviation (n=4).

Means with the same letter are not significantly different at the 5% level by Scheffé's F test.

DAS: Days after sowing.

Fig. 11 Root development of soybean at the harvest (n=3)
**Fig. 12** Changes in the number of tiller

- **O**: PG0
- **△**: PG100
- **□**: PG200
- **●**: PG500

**Days after transplanting**
- Basal dressing only
- Basal dressing + Topdressing

**Fig. 13** The ratio of different types of rice roots

- Red root
- Slightly black root
- Partial black root
- Black root
- Rotted root

**Fig. 14** Transverse sections' of rice roots stained by Evans blue

Transverse sections through the maturation zone in upper node of crown roots.

**Fig. 15** Comparison of brown rice yield

- Basal dressing only
- Basal dressing + Topdressing

*Fisher's LSD (<0.05)*
論文審査結果要旨

リン酸は高等植物の生育にとって三大要素の1つとして重要視されている。とりわけリン酸固定能の大きな火山灰土壌が多い我が国では農業にとっては栄養分というより土壌改良剤として使われてきた。本論文で対象とするリン酸石膏はそのリン酸製造過程でリン酸の約5倍量が副生する。リン酸石膏は、我が国では石膏ボードとしての使用が認められているが、諸外国では含まれる微量の放射性元素のため、農業以外の利用は禁止され、山積みされている。そこで申請者はわが国の主要な耕地土壌である沖積土と黒ボク土におけるリン酸石膏の土壌反応性と農業利用を検討した。

まず、第1に、沖積土と台地土に対するリン酸石膏の反応性を検討し、リン酸石膏施用量に応じて土壌pHが低下すること、土壌pHと土壌の一定荷電には負の相関があること、土壌溶液Alの増加量は交換性Alの1割以下であることを明らかにした。

第2に、リン酸石膏や炭カルなどの石灰資材の施用効果を検討し、土壌のCa可給度は、従来用いられてきた交換性Ca量より、水溶性Caの方が適切であることを明らかにした。またリン酸石膏施用に伴う水溶性Caの増加は、非アロフェン質黒ボク土で顕著であり、水溶性Ca量は、アロフェン含有や腐植含有量とも密接に関係することを明らかにした。

第3に、リン酸石膏のメロン育苗培土への施用効果を検討し、メロン根の発生や伸長促進に効果があることを明らかにした。またキュウリ根についても、Ca栄養や根張りの改善に顕著な効果があることを明らかにした。

第4に水田転換畑におけるダイズ栽培の問題点を明らかにし、石膏施用によって、沖積土の透水性や保水性の改善あるいはCa可給度の増強を通じて、根粒の着生、乾物生産が促進されることを明らかにした。

第5に、水稲栽培におけるリン酸石膏の影響を検討し、水稲の初期生育、分ゲツ促進、ひいては玄米収量向上に顕著な効果が見られることを明らかにする反面、過剰施用は硫化水素による根腐れ発生の危険性があることを、根組織の着色度と精密な顕微鏡観察から明らかにした。

このように申請者は、我が国の代表的耕地土壌におけるリン酸石膏の反応性と植物のCa栄養改善、土壌チルスの改善を通じて、リン酸石膏が作物の収量性改善に大きく貢献することを明らかにした。これの知見や技術はリン酸石膏の農業的有効利用に大きく貢献するものであり、審査員一同は申請者が博士（農学）に倣すると判定した。