京都大学	博士 (工 学)	氏名	Hendra Adhi Pratama
論文題目	Comparison of Cs Adsorption Characteristics of Soils in Japan and Indonesia (日本とインドネシアの土壌における Cs 吸着特性の比較)		

This dissertation presents the comparison of Cs adsorption characteristics of soils in Japan and Indonesia.

Chapter 1: Describes the background that motivates this research to be carried out, the objectives and the systematics of this research. This chapter also describes the study area which was selected in two different climatic area, that are in Takizawa Research Forest, Iwate, Japan which has temperate climate and in PUSPIPTEK Botanical Garden, Serpong, Indonesia which has tropical climate.

Chapter 2: A detailed literature review related to the background theory and recent methodology regarding the adsorption characteristics in soil is presented in this chapter. This chapter explained the Radiocesium Interception Potential (RIP), as intrinsic soil parameter, that is used to measure the Frayed Edge Sites (FES) in clay mineral quantitatively. Furthermore, recent methods to describe the implication of competitive ion in the sorption of heavy metals in soils also presented in this chapter.

Chapter 3: This chapter shows the detail of comparison results of analysing physico-chemical properties of soils in deciduous and coniferous forest from Iwate (Japan) and soil from Serpong (Indonesia). Climatic condition had made impact to the pedogenic process of soil. Thus, the composition of clay minerals, organic matter, inorganic and other soil particulate between Iwate soil from temperate region and Serpong from tropical region are very variable. High temperature and precipitation in Serpong area leads soil to faster desilication and thus, reducing the ability for cation sorption, which results low cationic exchange capacity (CEC). However, the surface area of this soil was higher than that of Iwate soils. Higher surface area in this soil also contribute to the distribution coefficient of Cs in soil. The heating soil treatment have a great impact to the surface area and caused removing of illitic clay mineral as well as organic matter. The composition of mineral and organic matter in soil have strong contribution to Cs adsorption than the type of vegetation of soil sampling area.

Chapter 4: Describes the development of rapid RIP measurement method and its comparison with the conventional method. Development of RIP measurement was performed using Cs stable in the acidic solution (pH 4) with the bath temperature of 40°C. The result of new rapid RIP measurement was in the same range of magnitude order with the result from the conventional method. Using this new method, there is no significant difference in RIP value between 3 and 5 days of Cs contact time to the soil solution.

Therefore, the RIP of soil could be estimated faster. The carrier effect of Cs in the RIP measurement was also investigated using radioactive Cs (¹³⁴Cs with stable carrier and mixture of ¹³⁴Cs, ¹³⁶Cs and ¹³⁷Cs without stable carrier) and using stable Cs (¹³³Cs). The results of RIP from all soil sample were varied from 27.27 to 522.66 mmol kg⁻¹.

Chapter 5: Explains the kinetic and equilibrium isotherm of Cs adsorption in soil. These are two important basics in order to understand adsorption process. The objective of this chapter is to obtain adsorptive capacity which is within the domain of the equilibria and to understand the diffusional resistance which is the domain of kinetics. The method of experiment is based on the previous chapter. The kinetic rates of Cs adsorption onto soil mineral were evaluated using pseudo-first order kinetic rate and pseudo-second order kinetic rate model. Cs sorption kinetic rate of all soil were better described by the pseudo-second order kinetic rate with larger coefficient of determination (R²) of all soil samples than 0.99. It suggests that the considered sorption process takes place as chemisorption. Equilibrium isotherms of Cs sorption were analyzed using linier regression of Langmuir and Freundlich isotherm. The result shows that equilibrium isotherms were better described by Freundlich model with the coefficient of determination (R²) of 0.947 and 0.981 for Iwate top-soil from coniferous and deciduous forest-floor respectively. These results suggest that the Cs adsorption onto soil is occurred on the heterogeneous adsorption surfaces.

Chapter 6: Describes the influence of competitive ion sorption in mineral soil to the RIP value. K⁺ and NH₄⁺ were used as competitive ions in this study. The adsorptions of Cs were better described by Freundlich isotherm. This might be due to relatively high concentration of Cs that used in the experiments. The effect of pH indicates that the trends of Cs adsorption and the distribution coefficient (K_D) increased linearly with the pH of solution. At around neutral pH (pH 6-8), the values of Cs adsorbed in soil and K_D value were within the range called the zero point of charge. In this range of pH, the adsorbed H⁺ dissociates into solution and leaving the surface with no net charge. At lower pH values, the surface sites would become more positively charged due to dissolution of a portion of soil and thus the surface will express less attraction for adsorbing Cs⁺. The competition with hydrogen ions for exchange sites also caused decreases in sorption.

Chapter 7: Summarizes all the important findings and highlights the conclusion from the obtained results in Chapters 3, 4, 5, and 6. Furthermore, the recommendation for future research subjects are also given in this chapter.

本論文は、福島第一原子力発電所と同様な事故がインドネシアで発生した場合の放射性セシウムの環境中挙動を予測するため、セシウムの土壌への吸着特性を温暖気候帯に属する岩手県の森林土壌(以下、岩手土壌)と熱帯気候帯に属するインドネシア Serpong の森林土壌(以下、Serpong 土壌)とで比較を行ったものである。得られた主な成果は以下のとおりである。

- 1) 土壌の主たる放射性セシウム吸着サイトとしては、粘土や土壌有機物に起因する通常のイオン交換基(REC)と、イライトの割れ目に起因するフレイドエッジサイト(FES)が存在するが、FESの定量化のためにはCremers et al.によって導入された放射性セシウム捕捉ポテンシャル(RIP)が有効である。本研究ではまずRIPの迅速測定法として、pH4、40℃の条件下で測定する方法を検討し、放射性セシウムの代わりに安定セシウムとICP-MSによる測定法を用いることが可能であること、従来法では5日要していた吸着平衡に達する期間を3日程度まで短縮することが可能であることを示した。
- 2) 岩手土壌と Serpong 土壌の様々な土壌特性を比較し、一般に Serpong 土壌の方が岩手 土壌よりも有機物が少なく陽イオン交換容量が小さいこと、RIP も粘土成分が少ない Serpong 土壌の方が岩手土壌よりも小さいことを示した。しかし、RIP の値はともに 79 から 128 mmol/kg 程度の値であり、スメクタイト構造を有するモンモリロナイトの RIP 測定値 629mmol/kg よりもかなり小さいことを示した。
- 3) セシウムの吸着速度モデルとして疑似 1 次モデルと疑似 2 次モデルを比較し、両土壌ともに疑似 2 次モデルの方で良く表現できることを示した。また、平衡吸着量は両土壌ともに Freundlich 型吸着等温式で表すことができることを示した。さらに、大量のカリウムイオンとアンモニウムイオンが存在する環境下にあっては、比較的高濃度セシウムの吸着が Freundlich 型の競争吸着等温式で表すことができることを示した。

以上の結果は、インドネシアの Serpong 土壌が日本の岩手土壌よりも放射性セシウムの保持能力が小さいことを示すものであり、インドネシアにおいて原子力災害が発生した場合のリスク評価や今後の原子力政策策定に大きく貢献するものであって、学術上、実際上寄与するところが少なくない。よって、本論文は博士(工学)の学位論文として価値あるものと認める。また、平成31年3月25日、論文内容とそれに関連した事項について試問を行って、申請者が博士後期課程学位取得基準を満たしていることを確認し、合格と認めた。なお、本論文は、京都大学学位規程第14条第2項に該当するものと判断し、公表に際しては、令和2年3月31日までの間、当該論文の全文に代えてその内容を要約したものとすることを認める。