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主 論 文 の 要 旨

Radiocesium contamination in woody plants

-Absorption process and utilization of contaminated wood-

論文題目 (樹木における放射性セシウム汚染

-吸収プロセスと汚染材の利用-)

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論 文 内 容 の 要 旨

The Fukushima Daiichi Nuclear Power Plant (FDNPP) accident in March 2011 resulted in the release of large amounts of radiocesium (^{137}Cs , ^{134}Cs) into the Fukushima Prefecture. Because forests cover approximately 70% of the entire prefectural area and are highly efficient at intercepting gaseous and particulate contaminants, the radioactive contamination of forests and forest products and the long-term effects of radiocesium on the forest biota and humans are of great concern, especially for ^{137}Cs that has a long half-life and is persistently recycled in forest ecosystem. To develop effective decontamination methods and to predict the future contamination, knowledge about how ^{137}Cs was taken up by trees and how ^{137}Cs was distributed within trees is important. There are three potential pathways of ^{137}Cs absorption by trees, i.e., foliar absorption, bark absorption, and root uptake from soil. However, the respective contribution of each source to the ^{137}Cs detected in wood is difficult to evaluate. In particular, the possibility of bark absorption is questionable, due to that the outer bark consists of only dead tissue and a moisture-impermeable layer exists between the outer bark and inner bark. In addition, the magnitude of bark absorption, how ^{137}Cs moves within trees

after bark absorption, and whether differences exist in the absorption process among different tree species are also not well understood. The purposes of this study are to investigate the bark absorption of ^{137}Cs and its subsequent translocation and distribution in trees, as well as evaluating the possible utilization of ^{137}Cs -contaminated wood.

Firstly, we investigated the bark absorption and translocation of ^{137}Cs in three widely distributed tree species in Fukushima Prefecture: Japanese cedar (*Cryptomeria japonica*), Konara oak (*Quercus serrata*), and Red pine (*Pinus resinosa*). We determined the ^{137}Cs concentration in the outer bark, inner bark, sapwood, heartwood of disks collected from different heights, as well as in leaves/needles from tree top. In addition, because the chemical behavior of ^{137}Cs is expected to be almost identical to ^{133}Cs , analysis of ^{133}Cs is useful for understanding the behavior of ^{137}Cs in a forest ecosystem. We conducted a simulation experiments by applying $^{133}\text{CsCl}$ aqueous solution on the bark at 1.2 m-height above ground of these tree species, and we analyzed the ^{133}Cs concentration in each of the above-mentioned organs for both ^{133}Cs -treated trees and control trees. The results are as follows: 1) we confirmed that Cs could enter trees through bark for all the three tree species, but the infiltration ability differed among tree species. The amount of Cs entered trees was with the order of *C. japonica* > *Q. serrata*, *P. densiflora*. The infiltration ability was affected by the several factors that governing the bark absorption process, such as the fixation/adsorption of Cs by the outer bark, the mechanism and route for translocation of Cs through the outer bark to the inner bark, and the translocation from inner bark to sapwood, the process of which is not well understood and may depend on the interaction of phloem and xylem. 2) The bark-derived Cs exhibited

different radial distribution patterns, Bark-derived ^{133}Cs was translocated to heartwood of *C. japonica* and *P. densiflora*, however, it is uncertain whether ^{133}Cs in heartwood of *Q. serrata* originated from bark or not, because ^{133}Cs concentration in heartwood of *Q. serrata* was comparable between ^{133}Cs -treated trees and controls. ^{133}Cs tended to accumulate in heartwood of *C. japonica* while decreased towards heartwood of *Q. serrata* and *P. densiflora*. In the vertical direction, ^{133}Cs was translocated to tree top of *C. japonica*, but this phenomenon was not observed for *Q. serrata* and *P. densiflora* during our experimental period.

Secondly, we proposed the using of $^{133}\text{Cs}/\text{K}$ and $^{137}\text{Cs}/^{40}\text{K}$ ratios in wood as evidence of bark absorption. We hypothesized that: because the natural ^{133}Cs , K, and ^{40}K are absorbed only from the soil, the radial distribution of $^{133}\text{Cs}/\text{K}$ ratio should be homogenous and that no discrimination of Cs and K is occurring via root uptake and translocation in the wood. Therefore, if ^{137}Cs is absorbed only from the soil, the distribution of $^{137}\text{Cs}/^{40}\text{K}$ in the wood should be similar to that of $^{133}\text{Cs}/\text{K}$, i.e., show a homogenous pattern in the radial direction. Based on this hypothesis, we determined the $^{133}\text{Cs}/\text{K}$ and $^{137}\text{Cs}/^{40}\text{K}$ ratio in wood of the studied tree species, the results demonstrated the contribution of bark absorption to ^{137}Cs in wood of *C. japonica* and *Q. serrata*. In addition, we also found the absorption ability of ^{137}Cs in *P. densiflora* was low, not only for bark absorption, but also for root uptake and foliar absorption.

Finally, we investigated the utilization of ^{137}Cs -contaminated wood, according to the above-mentioned results, we confirmed that part of Cs in wood exists in the ionic form, which means that Cs could be removed through chemical treatments. Kraft pulping was conducted for ^{137}Cs contaminated and ^{133}Cs treated *C. japonica* wood and bark, and the behavior of Cs during the kraft pulping process was

examined. The results showed for wood samples, most of Cs was transferred to black liquor, while only a minor amount ($< 1\%$) of Cs was present in the wood pulp, which suggested ^{137}Cs contaminated *C. japonica* could be used in pulp and paper industries. In the case of bark samples, although the majority of Cs was present in the black liquor, the proportion of Cs in the bark pulp was much higher than that in the wood pulp. In addition, the Cs in the precipitation of the bark was higher than that in the wood, possibly because the Cs in the bark was combined with some components, which is insoluble in alkaline solution. The experiment was conducted for *C. japonica*, but it is expected that the results are also applied for *Q. serrata* due to that the main chemical composition is similar between *Q. serrata* and *C. japonica*. In addition, giving the low ^{137}Cs concentration in wood of *P. densiflora* and the low absorption ability of Cs in *P. densiflora*, we speculated that the wood of *P. densiflora* can be used as usual, at least in the forests where we collected the samples.

In conclusion, we confirmed the bark absorption of Cs in *C. japonica*, *Q. serrata*, and *P. densiflora*. The infiltration ability through bark differed among trees species. The applied ^{133}Cs exhibited different radial and vertical distribution patterns among tree species. The ^{137}Cs -contaminated wood could be used in pulp and paper industries. These findings suggested that bark absorption is an important contribution to tree contamination, at least in the early stage of accident, and the contaminated forest is still valuable if it is properly used.