RADIONUCLIDE UPTAKE AND LONG TERM BEHAVIOR OF Cs-137, Cs-134 AND K-40 IN TREE RINGS OF SPRUCE

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The uptake and the long term behavior of Cs-137, Cs-134 and K-40 in the annual tree rings of spruce were examined. The youngest tree rings which are most active in water transport have higher activity concentrations of K-40 and of radioesium than the older ones. The activity concentration of Cs-137 in a water transporting tree ring can be well described as a function of the activity concentration of K-40. Furthermore a depth profile of the soil was taken and gives information about the depth distribution of radioesium and K-40.

Radioactive fallout from nuclear weapon tests and accidental releases of radionuclides from nuclear power plants led to a contamination of wide areas on the northern hemisphere. High activity concentrations are still found in forest ecosystems which decrease only very slowly. Tree ring examinations are a useful means to study the distribution pattern of artificially and naturally occurring radionuclides. A comparison to the fallout history is illuminating for transport phenomena in spruce. KOHNO et al. and KUDO et al. found that the distribution of radioesium along the tree rings of Cryptomeria japonica D. is not correlated with that of fallout deposition; Cs-137 was found in tree rings formed earlier than 1945. The object of our work is the determination of the tree ring distribution of total radioesium and K-40 and the relation Cs-137 to K-40 in a 85 year old spruce tree (Picea abies L. Karst.).

Experimental

Sample description: The tree studied (Picea abies L. Karst.) was 85 years old and grew up in southeast Germany. In spring of 1990 the tree was thrown over by a storm. The mean value of temperature for the last 30 years was about 8°C and the mean value of precipitation was about 830 mm. The stem was cut into several disks with 5 cm thickness. After the bark was removed the surfaces were planed to avoid any contaminations. Now the disks were separated into single tree rings with a chisel. Some thin tree rings and the inner ones had to be pooled to get enough mass for detection. The tree ring masses range from 0.4 kg to 0.9 kg dry weight. The samples were dried at 105°C and then ashed in a muffle furnace at 450°C. After ashing the reduced sample volumes vary between 1.5 ml and 4.0 ml which causes an increased counting efficiency for gamma spectrometry. The depth profile of soil represents a mixture of 16 single depth profiles taken within the root zone of the spruce. A drill with 2 cm diameter was used to take the soil samples. The first 10 cm always 1 cm soil layers were collected, from 10 cm to 24 cm depth only 2 cm soil layers were taken. The soil layers were dried at 105°C and sieved with 2 mm mesh width. The volume ranged from 25 ml to 70 ml.
Gamma spectrometry: The activity concentrations were determined employing gamma spectrometry with a HPGe-detector (coaxial geometry, 250 cm$^3$ active volume, 10 cm low-level lead shielding). Volume dependent calibrations were performed for LSC-vial geometry and a 100 ml polyethylene box which were used for detection. Quality assurance was guaranteed by regular participation in comparative measurements of radioactive samples issued by the Bundesgesundheitsamt, furthermore internal quality assurance measurements were performed monthly. At a mean counting time of 4000 min the detection limit (confidence level: 95%, LSC-vial geometry) is 7 mBq/sample for Cs-137 and Cs-134 and 80 mBq/sample for K-40 (according to KTA-rule 1504 of German regulatory authorities).

The following definitions are used in the succeeding text:

- activity concentration of a nuclide: $a_{\text{nuclide}}$
- Cs-137 from Chernobyl fallout: $a_{\text{Cs-137Ch}}$
- Cs-137 from nuclear weapon fallout: $a_{\text{Cs-137nw}}$

As the ratio of Cs-137 to Cs-134 from Chernobyl fallout is well known and the concentration of Cs-134 from the nuclear weapon fallout is negligible the proportion of Cs-137 from Chernobyl fallout is calculated as:

$$a_{\text{Cs-137Ch}} = 1.76 \cdot a_{\text{Cs-134}}$$

This ratio was found in southeast Germany at the time of the Chernobyl accident. To calculate the proportion of Cs-137$^{\text{Ch}}$ the activity concentration of Cs-134 has to be corrected due to radioactive decay and multiplied with the factor 1.76. Therefore the activity concentration of Cs-137$^{\text{Ch}}$ is strongly correlated with the activity concentration of Cs-134. To calculate the activity concentration of Cs-137$^{\text{nw}}$ the portion of Cs-137$^{\text{Ch}}$ has to be subtracted from the total Cs-137 activity concentration. This method has the advantage that the contributions to Cs-137 can be separated according to their origin.

The statistical treatment of the relation Cs-137 to K-40 was executed using the linear correlation coefficient $r$. This correlation coefficient $r$ is calculated according to

$$r = \frac{n \cdot \sum x \cdot y - \sum x \cdot \sum y}{\sqrt{[n \cdot \sum x^2 - (\sum x)^2] \cdot [n \cdot \sum y^2 - (\sum y)^2]}}$$

with

- $n$: number of samples
- $x$: activity concentration of K-40
- $y$: activity concentration of Cs-137

The correlation coefficient $r$ ranges from +1 to -1. A positive correlation results from $x$ and $y$ increasing, a negative correlation from increasing $x$ and decreasing $y$. The dependence between $x$ and $y$ is more probable the nearer $r$ is to the +1 or -1. Weak dependence valids for $r$ ranges around 0. The error depends on the number of samples. At the same value of the correlation coefficient the error becomes smaller by an increasing number of samples.

Results and Discussion

In Figure 1 the activity concentrations of K-40, Cs-137$^{\text{nw}}$ and Cs-137$^{\text{Ch}}$ (representing Cs-134) are plotted versus the year of growth. In the youngest tree rings which are most active in water transport higher activity concentrations of radiocesium and of K-40 are detected than in the older ones. The radial distribution of radiocesium is similar to that for