Impact of afforestation on long term $^{137}$Cs and $^{90}$Sr recycling from a waste burial in the Chernobyl Red Forest

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INTRODUCTION

Following the Chernobyl reactor fire in April 1986, acute irradiation caused complete decay of pine trees in forest located in direct vicinity of ChNPP. The so-called "Red Forest", a 1500 ha of Scots pine stand where the absorbed doses values exceeded the destructive value for coniferous species (> 60 Gy), is still today one of the most contaminated terrestrial ecosystems on Earth. Emergency clean-up activities consisted of in situ burial of contaminated topsoil layers and dead trees in about two hundred sub-surface trenches involving 1 million m$^3$ of radioactive materials. The trenches of Red Forest waste dump were then covered with 20-30 cm layer of "clean" sand and the site was revegetated with a mixture of pine, birch and bushes to prevent secondary contamination due to soil erosion or wind resuspension (Kozubov and Taskaev, 2002).

Large uncertainties are associated with the functioning of that improvised repository which remains a source of radionuclides (RN) dispersion into the environment. In particular, the role of new forest plantations as a long term stabilization factor of the radioecological situation in the afforested waste burial zone is questioned. With forest growth, future risk of radioactivity dissemination may involve the root uptake of bio-available radionuclides from deep layers in addition to the leaching of RN with downwards water fluxes. Our investigation aimed to assess the fraction of activity which could be extracted from the trenches due to root uptake by trees and further involved in natural biological cycles. For that purpose, we compared the transfer of $^{90}$Sr and $^{137}$Cs to 15 years-old Scots pine trees ($Pinus sylvestris$ L.) growing at the Chernobyl Pilot Site, directly on the trench no.22 and outside the trench.

MATERIALS AND METHODS

Site description

The trench no.22 (~70 m long, 9 m wide and 2.5 m deep) is located in the Chernobyl exclusion zone, in the middle of the Red Forest, approximately 2.5 km South-West of the ChNPP unit 4. In the frame of the IRSN-UIAR-IGS projects EPIC, a specific area of 0.8 ha., the Chernobyl Pilot Site (CPS) was delineated around the trench no.22 and further equipped for in situ radioecological monitoring (Dewiere et al., 2004). In the 30-km Chernobyl zone, the density of soil contamination with $^{90}$Sr, $^{137}$Cs, $^{154}$Eu, $^{238}$Pu, $^{239+240}$Pu and $^{241}$Am was clearly related to the deposition of fine-dispersed fuel particles of the Chernobyl fallout (Kashparov et al., 2003). Waste material of the trench no.22 is logically characterized by a similar series of radionuclides. Bugai et al. (2004) estimated the total inventory of the radioactivity in trench no.22 as 600 ± 240
GBq for $^{137}\text{Cs}$ and $290 \pm 140$ GBq for $^{90}\text{Sr}$. While the vertical distribution of $^{90}\text{Sr}$ and $^{137}\text{Cs}$ activities was rather heterogeneous, it well reflected the presence of waste material from 30 till 270 cm in depth. Outside the trenches, RN profiles with significant activities restricted to surficial soil layers (0-50 cm) mainly result from residual initial contamination and processes of litterfall and further downward migration of the leached radionuclides. In absence of a systematic monitoring, we assumed that surface soils outside the trench were contaminated with a $^{137}\text{Cs}/^{90}\text{Sr}$ activity ratio similar to that of the waste material.

**Vegetation sampling and analysis**

According to the singular non-uniform spatial structure of the pine plantation, our best estimates of average pine density was 1500 trees/ha. The distribution of trunk circumferences at 1.30 m (breast height, CBH) was determined from a random measurement of about 160 pine trees located within the CPS area. Ten average trees were used to measure the mean average height of the stand. Detailed information on the annual average biomass increment of the different tree compartments as well as on litterfall production was then chosen in biomass tables established for Scots pine in the study area. Basic mensurational data indicated that, compared to a traditional pine plantation of the same age, the current pine stand was characterized by a wider range of tree sizes with less high and more large trees. We estimated however that important generic development criteria (e.g. mean stem volume, shape coefficient, …) for a reference pine stand of productivity class I best matched those of our inventory. For tree sampling, we distinguished two groups: the pines growing directly on the trench no.22 (tree IN with a deep soil contamination involving waste materials) and outside the trench no.22 (tree OUT with a residual surface soil contamination). For each tree group, nine trees were chosen in the different CBH categories, proportionally to their relative importance. The different samples were compartmented into major annual and perennial parts: stemwood, inner and outer bark, needles of the year and >1y., twigs and branches. On the basis of annual biomass production and respective average element concentrations in the various tree compartments, the biological cycling dynamics of $^{90}\text{Sr}$ and $^{137}\text{Cs}$ in the ecosystem was quantitatively described in terms of annual fluxes: total incorporation, net uptake from soil, immobilization in woody organs and growing foliage biomass, return to the soil and internal transfer. The terms used to assess the element cycling in pine plantation as well as detail of the calculation were described elsewhere by Thiry et al. (2005). Activity levels in representative sample of each tree organs were determined after calcination at 550°C for 24 h, followed by wet acid digestion of the ashes. The $^{137}\text{Cs}$ activities in the final digests were measured by means of $\gamma$-spectrometry (ADCAM-300 with the GEM-30185 detectors, EG&G ORTEC, USA). The $^{90}\text{Sr}$ activities were measured with a $\beta$-spectrometer SEB-001 (Ukraine) after application of a standard radiochemical procedure to extract $^{90}\text{Sr}$ from the samples.

**RESULTS AND CONCLUSIONS**

For each tree group, activity level of radiostrontium in nearly all pine compartments exceeded that of radioceasium by about one order of magnitude. Despite different activity levels and growing conditions between tree groups due to the subsoil, the respective allocation pattern of $^{137}\text{Cs}$ and $^{90}\text{Sr}$ among pine components was similar between sites. Logically, $^{137}\text{Cs}$ as an analogue of potassium was shown to concentrate in physiologically active organs, the last-formed needles and twigs in particular, while $^{90}\text{Sr}$ as an analogue of calcium accumulated more importantly in
ligneous components and senescing foliage. Today, about 82 % of the total \(^{90}\)Sr content in aboveground biomass and 54 % of \(^{137}\)Cs was accumulated in woody organs including bark, the rest being incorporated in the foliage compartment.

After 15 years of growth, an average tree IN i.e. growing on the trench had accumulated 0.70 MBq of \(^{137}\)Cs and 35.09 MBq of \(^{90}\)Sr in standing biomass. The aboveground biomass of tree IN was contaminated with 1.66 times more \(^{137}\)Cs than that of tree OUT and 5.32 times more \(^{90}\)Sr. These differences in tree contamination between sites provided evidence that part of the trench contamination is translocated upward due to root uptake and further accretion in tree biomass. The total amount of radioactivity already recycled due to biomass turnover was however most likely underestimated since our inventory of radioactivity was restricted to standing biomass and did not take into account of the element accumulated in the forest floor. For a realistic view of the current radionuclide dynamics, the main processes governing the \(^{90}\)Sr and \(^{137}\)Cs biological cycling were quantitatively described in terms of annual fluxes as depicted in Table 2. Our calculations led to the following observations:

**Table 1.** Annual fluxes depicting the \(^{137}\)Cs and \(^{90}\)Sr biological cycling in plantation of pines growing out of the trench no.22 (Tree OUT) and directly on the trench (Tree IN)

<table>
<thead>
<tr>
<th>Fluxes</th>
<th>(^{137})Cs (MBq ha(^{-1})y(^{-1}))</th>
<th>(^{90})Sr (MBq ha(^{-1})y(^{-1}))</th>
</tr>
</thead>
<tbody>
<tr>
<td>Incorporation</td>
<td>303</td>
<td>513</td>
</tr>
<tr>
<td>Uptake (1+2+3)</td>
<td>135</td>
<td>164</td>
</tr>
<tr>
<td>(1) Immobilization</td>
<td>21</td>
<td>26</td>
</tr>
<tr>
<td>(2) Return to the soil</td>
<td>96</td>
<td>111</td>
</tr>
<tr>
<td>(3) ?f</td>
<td>18</td>
<td>26</td>
</tr>
<tr>
<td>Internal transfer</td>
<td>176</td>
<td>363</td>
</tr>
</tbody>
</table>

- The flux of \(^{137}\)Cs incorporated in new organs amounted to 303 and 513 MBq ha\(^{-1}\)y\(^{-1}\) for tree OUT and tree IN, respectively. The internal transfer processes, especially from the senescing foliage, was a major source of \(^{137}\)Cs translocation to new biomass production. The net root uptake contributed in fact to only 40 % of the current \(^{137}\)Cs incorporation rate. Litterfall and throughfall return to the soil a large part of the \(^{137}\)Cs uptake (~70 %), resulting in a limited \(^{137}\)Cs immobilization in perennial compartments (~15 % of annual uptake) which is in agreement with Cs dynamics in young pine plantation as previously observed by Goor and Thiry (2004).

- For \(^{90}\)Sr, we measured an annual net uptake of 3476 and 17057 MBq ha\(^{-1}\)y\(^{-1}\) for tree OUT and tree IN, respectively. For each tree group, the \(^{90}\)Sr root uptake was greater than its incorporation into new biomass. Current uptake rate exceeded that of allocation to new organs by about 70 %. It has been shown that strontium follows calcium metabolism fairly closely. Ca is hardly retranslocated by the phloem sap resulting in high accumulation in senescing foliage and woody tissues. Besides a "luxury" consumption of Ca is common for forest stand (Van der Stegen and Myttenaere, 1991). The negative value observed for \(^{90}\)Sr internal transfer, especially from the foliage, is consistent with the strategy developed by trees to counteract the excessive uptake of Ca. On the other hand,
the fraction of $^{90}$Sr uptake which was immobilized in woody compartments (~30 %) was twice as high as that of $^{137}$Cs (~15 %), confirming also that stemwood act as a durable sink for the $^{90}$Sr moved from the roots.

For tree IN, our results showed that the current element cycling dynamics involve a potential of pine to annually extract up to 0.33 % of the $^{90}$Sr pool in the trench and only 0.0015 % of that of $^{137}$Cs. On the trench, the extraction rate of $^{90}$Sr by vegetation is then 220 times more efficient than that of $^{137}$Cs. That large flux of $^{90}$Sr recycling is at least of the same magnitude than $^{90}$Sr downward migration losses as reported by Bugai et al. (2002). That demonstrates that forest tree uptake can induce and prolong a significant secondary contamination of surface soils covering the trenches of the Red Forest. Very interesting was also the difference noted in $^{90}$Sr/$^{137}$Cs ratio between tree IN and tree OUT based on net annual uptake rates. Our results indicated that with reference to $^{137}$Cs, $^{90}$Sr was 4.1 times more available to recycling by tree IN than by tree OUT. That observation suggests a preferential $^{90}$Sr uptake from deeper layers while pine roots take up $^{137}$Cs preferentially from surface layers. Regardless of the cause of the differences in recycling rate for the various RN, these differences have consequences for the long term modelling of the RN pool contained by trenches of the afforested Red Forest waste dump.

REFERENCES