Long-term cycling of $^{137}$Cs in *Pinus Kesiya* at Wat Chan Thailand

T Itthipoonthanakorn¹ and G Shaw²

¹Office of Atoms for Peace, 16 Vibhavadi Rangsit Road, Bangkok 10900, Thailand
²School of Biosciences, University of Nottingham, Sutton Bonington, LE12 5RD, UK

Email: thawatchai.i@oap.go.th

Abstract. Surface soil and above-ground parts of *Pinus kesiya* (Three-needled pine) were collected at Wat Chan pine forest to study cycling of $^{137}$Cs. Cs-137 derived from nuclear weapons tests up to 1963 was found to have accumulated at a shallow depth (<5 cm) in the forest soil. Calculated total deposition of $^{137}$Cs was in the range of previous studies and could be used as a reference for nuclear test deposition of $^{137}$Cs fallout in northern Thailand and nearby locations. Potassium in soil does not appear to be a good indicator for long-term radiocaesium cycling. Finally, ‘available’ stable caesium ($^{133}$Cs) in the soil was found to be an appropriate indicator for studying uptake of $^{137}$Cs by *Pinus kesiya* at Wat Chan.

1. Introduction

Forests are important receptors of atmospheric pollutants, including fallout from nuclear explosions and nuclear facility accidents [1]. Coniferous forest is a more effective interceptor of atmospheric fallout than any other natural and semi-natural ecosystem [2]. Major radioactive contamination of forest ecosystems occurred after the Kyshtym accident (1957), during the era of atmospheric nuclear weapons testing which peaked in 1962, and following the accidents at the Chernobyl and Fukushima nuclear power plants in 1986 and 2011, respectively [3]. Among the radionuclides released by both reactor accidents and nuclear weapons to the environment $^{137}$Cs is one of the most important and often dominates the doses to humans during the medium- and long term after initial deposition [4].

After the Chernobyl accident, radioactive fallout measurements across the region of East Asia showed mostly undetectable deposition of radiocaesium [5], [6]. Insignificant contribution of $^{137}$Cs from Chernobyl from 1986 to 1990 was found throughout Vietnam [7] where a ‘latitude mean deposition density’ ranging from 237 Bq m$^{-2}$ at 16° S latitude to 1,097 Bq m$^{-2}$ at 16° N originated from nuclear weapons testing [8]. Deposition of $^{137}$Cs from the Fukushima accident was negligible in the region: a transient peak air concentration of 37 μBq m$^{-3}$ in Ho Chi Minh City was reported [9].

Knowledge on migration of $^{137}$Cs in forest ecosystem pathways is needed to quantify the radiation dose which humans receive as well as to explore and evaluate any proposed remedial and abatement strategies. A number of publish papers have been conducted in temperate forest ecosystems whilst few have done in tropical forest ecosystems.

Data of $^{137}$Cs cycling in pathways of forest ecosystem is needed to quantify the radiation dose which humans receive as well as to explore and evaluate any proposed remedial and abatement strategies. Transfer factor for prediction of $^{137}$Cs distribution in forest ecosystems have been investigated mostly in temperate forest [2]. The uptake of the alkali metals (K, Rb and Cs) by spruce needles (as measured by concentration ratios, CRs) should be similar, but large differences are
observed due to their different sorption behaviour in the soil [10]. CRs for $^{137}$Cs should be closer to those for exchangeable $^{133}$Cs than for total $^{133}$Cs, which is largely in insoluble mineral forms in most soils. Consequently, fallout $^{137}$Cs and exchangeable $^{133}$Cs are more available for plant uptake than total native $^{133}$Cs [11].

However, in tropical forests, specific climatic and environmental conditions such as dry and wet periods and high rates of weathering, may perhaps modify $^{135}$Cs behavior. Therefore, relevant research in tropical forest of Thailand is needed for establishing of suitable remedial and abatement strategies to prepare for future impact of radionuclide contamination from nuclear power and research programs in Thailand and neighboring countries.

In this study, long-term behavior of $^{137}$Cs, attributed to atmospheric nuclear weapons tests in 1950-60s, was investigated both in surface soil and pine trees in a tropical forest in northern Thailand.

Wat Chan forest, located has been selected due to their location in northern part of Thailand which received more deposition of nuclear fallout than southern part. Uptake of $^{137}$Cs into Pinus Kesiya is compare to of pine tree available in IAEA data [2].

2. Methodologies

2.1. Sampling location

Wat Chan Pine forest is a native tropical pine forest of Pinus kesiya (Three-needled pine) located in Kanlayani Watthan district of Chiang Mai Province in northern Thailand (19° 4′N 98° 18′E). The soil beneath the forest is a red-yellow podzolic type. Sampling at Wat Chan forest was conducted in February 2016.

2.2. Soil sampling and treatment

Six soil cores (6.8 cm diameter and 13 cm length) were sampled using a steel soil corer. Six soil cores sampling randomly by walking across an area of approximately 1 hectare is considered adequate number to represent soil core in this forest. Soil cores were cut into 1 cm layers with respect to depth then ground and oven dried at 105°C overnight.

2.3. Tree core and needle sampling and treatment

A screw-type increment tree corer was used to take two narrow (5 mm diameter) cores of wood across one half of the width of a representative pine tree as close as possible to the corresponding soil sampling point. The tree cores were taken approximately 40 and 60 cm above ground level. Finally, living (green) pine needles were collected as near as possible to each soil sampling point. Tree core and needle samples were dried overnight at 105°C then ground to a fine powder in an ultra-centrifugal mill.

2.4. Measurement of $^{137}$Cs

Dried and ground soil samples were packed into plastic Petri dishes (3.5 cm diameter, 1.1 cm height). The internal volume of the Petri dish was completely filled with sample and the dish plus loose-fitting lid were sealed using Parafilm®, final bulk density were between 0.9 – 1.4 g cm$^{-1}$. Soil samples were counted for 24 hours using a high resolution gamma-ray spectrometry system with a high-purity germanium (HP-Ge) detector to determine the activity concentrations of $^{137}$Cs at 661.7 keV, in soil samples. A mixture of sulfuric acid washed sand (undetectable background of $^{137}$Cs) with a solution of a certified mixed gamma standard (including 587 Bq kg$^{-1}$ of $^{137}$Cs) obtained from the National Physical Laboratory (R08-01-2016040167-1) is used for efficiency calibration (final bulk density is 1.3 g cm$^{-1}$).

2.5. Measurement of stable Cs and competitive elements

Activity concentrations of $^{137}$Cs in tree cores and needles were undetectable by gamma-ray spectrometry. Therefore, as a surrogate, stable caesium ($^{133}$Cs) was used to evaluate long-term cycling of caesium isotopes in undisturbed forest ecosystems. Samples of soil, tree cores and needles of Pinus


kesiya were extracted using HNO₃ and analysed using ICP-MS to determine concentrations of ¹³³Cs and the alkali metal ‘competitor’ elements potassium and rubidium. In addition, soil was extracted with NH₃NO₃ to determine the available fractions of ¹³³Cs, K and Rb.

2.6. Calculation of transfer factors of stable Cs and competitive elements

Migration of stable caesium and competitor elements from soil to wood (tree cores), needles and bark of Pinus kesiya was assessed using aggregated transfer factors and concentration ratios. Since ¹³³Cs was detectable only within the upper 6 cm depth of soil these calculations are based on deposition of stable caesium, potassium and rubidium distributed in the upper 6 cm depth of the forest soil. Equations 1 and 2 were used to calculate aggregated transfer factors and concentration ratios. Transport of alkali metals from wood (tree cores) to needles was quantified using a translocation factor, shown in Equation 3.

Aggregated transfer factor

\[ T_{ag} = \frac{AP}{TD} \]  

Where \( T_{ag} \) = Aggregated transfer factor (m² kg⁻¹)
\( AP \) = Activity concentration in forest vegetation compartment (Bq kg⁻¹)
\( TD \) = Total deposition to forest floor within upper 6 cm depth of soil profile (Bq m⁻²)

Concentration ratio

\[ CR = \frac{M_P}{M_S} \]  

Where \( CR \) = Concentration ratio
\( M_P \) = Mass concentration in plant (Bq kg⁻¹ dry or fresh weight)
\( M_S \) = Mass concentration in soil within upper 6 cm depth of soil profile (Bq kg⁻¹ dry weight)

Translocation factor

\[ f_u = \frac{M_S}{M_T} \]  

Where \( M_N \) = Mass concentration in needle (mg kg⁻¹ dry weight)
\( M_T \) = Mass concentration in wood (mg kg⁻¹ dry weight)

2.7. Total deposition of ¹³⁷Cs

2.7.1. Deposition inventory of ¹³⁷Cs in soil

The detectable ¹³⁷Cs was found to a soil depth of 6 cm. The total deposition inventory of ¹³⁷Cs in soil, therefore, was calculated assuming that negligible ¹³⁷Cs had migrated below 6 cm depth in soil cores.

2.7.2. Deposition of ¹³⁷Cs in wood and needles of Pinus kesiya

As well as the deeper depths of soil, ¹³⁷Cs in tissue of Pinus kesiya was undetectable. Estimates of activity concentrations of ¹³⁷Cs in wood (tree cores) and needles of Pinus kesiya were made using the geometric mean aggregated transfer factor of pine (\( 1.7 \times 10^{-3} \) and \( 1 \times 10^{-2} \) m² kg⁻¹ dry weight, respectively) provided by IAEA [2]. Calculated activity concentrations were converted to deposition inventory of ¹³⁷Cs using calculated biomass.

2.8. Biomass calculation

Biomass of Pinus kesiya was estimated using a biomass equation for a single tree [12] and the average number of standing Pinus kesiya trees per unit area in 6 replicate areas of 50 × 50 m surrounding or adjacent to the soil sampling sites. Biomass of needles and branch compartments of a single tree were calculated using Equations 4 and 5.
WB = 0.0012 x (D^2 x H)^{1.0996} \quad (4)
WL = 0.4536 x (WB)^{0.7933} \quad (5)

When
WB = Biomass of branch (kg)
WL = Biomass of needles (kg)
D = Diameter of trunk at breast height (cm)
H = Height of tree (m)

3. Results and discussions
3.1. Vertical distribution of $^{137}$Cs in soil
Vertical distributions of $^{137}$Cs in forest soil are shown in Fig.1. The shallow distribution of $^{137}$Cs is indicated with almost all $^{137}$Cs distributed within upper the 6 cm of the soil core. Half-depth and mean migration rate of $^{137}$Cs in collecting year (2016) were approximately 2 cm and 0.38 mm y$^{-1}$, respectively.

![Figure 1](image_url)

**Figure 1.** Depth profiles of average $^{137}$Cs (a), $^{137}$Cs deposition inventories (b) and percentage $^{137}$Cs inventory (white circle) and accumulated percentage $^{137}$Cs inventory (dark circle) (c), decay corrected to 1963, in soil collected at Wat Chan (6 replicates, the horizontal bars are standard errors of means).

3.2. Uptake of stable Cs into wood and needles of Pinus kesiya
Aggregated transfer factors and concentration ratios for stable caesium, potassium and rubidium (Tables 1 and 2) suggest that stable caesium is less preferentially absorbed than potassium and rubidium during uptake into wood and needles of *Pinus kesiya*.

Because stable caesium is largely in insoluble mineral forms in most soils [11] aggregated transfer factors calculated using total concentrations of stable caesium are lower than the range of $T_{ag}$ for radiocaesium in wood and needles of pine suggested by IAEA [2]. However, $T_{ag}$ values calculated using available concentrations of stable caesium are consistent with IAEA data ($1.1 \times 10^{-4}$ and $2.1 \times 10^{-2}$ m$^2$ kg$^{-1}$ for pine wood; $2.4 \times 10^{-4}$ and $9.2 \times 10^{-2}$ m$^2$ kg$^{-1}$ for pine needles) [2] and could provide an analogue for radiocaesium when studying $^{137}$Cs cycling in forest ecosystems of Wat Chan.
Table 1. Aggregated transfer factors ($T_{ag}$) of $^{133}$Cs, $^{39}$K and $^{85}$Rb in wood (n = 6) and fresh needles (n = 6) in Pinus kesiya calculated on a dry weight basis for wood and needles using total and available concentrations in the upper 6 cm of the soil (n = 6).

| Soil fraction   | Tree tissue | $^{133}$Cs | $^{39}$K | $^{85}$Rb |
|----------------|-------------|------------|----------|-----------|
| Total concentration | Wood        | $1.96 \times 10^{-5}$ | $1.50 \times 10^{-2}$ | $1.16 \times 10^{-3}$ |
|                  | Fresh needles | $8.72 \times 10^{-5}$ | $6.37 \times 10^{-2}$ | $3.22 \times 10^{-3}$ |
| Available concentration | Wood        | $3.24 \times 10^{-4}$ | $2.29 \times 10^{-3}$ | $5.52 \times 10^{-3}$ |
|                  | Fresh needles | $1.44 \times 10^{-3}$ | $9.74 \times 10^{-2}$ | $1.54 \times 10^{-2}$ |

Table 2. Concentration ratios (CR) of $^{133}$Cs, $^{39}$K and $^{85}$Rb in wood (n = 6) and fresh needles (n = 6) in Pinus kesiya calculated on a dry basis for wood and needles using total and available concentrations in the upper 6 cm of the soil (n = 6).

| Soil fraction   | Tree tissue | CR (dry weight basis) |
|----------------|-------------|-----------------------|
| Total concentration | Wood        | $1.65 \times 10^{-3}$ | $1.26$ | $9.74 \times 10^{-2}$ |
|                  | Fresh needles | $7.36 \times 10^{-3}$ | $5.34$ | $2.71 \times 10^{-4}$ |
| Available concentration | Wood        | $2.76 \times 10^{-4}$ | $1.95$ | $4.71 \times 10^{-4}$ |
|                  | Fresh needles | $1.23 \times 10^{-1}$ | $8.31$ | $1.31$ |

Translocation factors calculated for stable caesium and potassium (Table 3) suggest that, once stable caesium and potassium have been taken up from the soil by Pinus kesiya, the relative partitioning of these two elements between wood and needles is comparable.

Table 3. Wood-to-needle translocation factors ($f_{tr}$) for $^{133}$Cs, $^{39}$K and $^{85}$Rb in Pinus kesiya calculated on a dry weight basis.

| Translocation factor ($f_{tr}$, dry weight basis) |
|----------------|----------|----------|
| $^{133}$Cs | $^{39}$K | $^{85}$Rb |
| 4.45         | 4.25     | 2.78     |

3.3. Standing biomass of Pinus kesiya

Table 4. Density and standing biomass of Pinus kesiya at Wat Chan.

| Tree density (m$^2$) | Biomass (kg m$^{-2}$) |
|---------------------|----------------------|
|                     | Timber | Branches | Needles | Total |
| 2.52 $\times 10^{-2}$ | 15.9 ± 3.18 | 4.27 ± 1.06 | 0.655 ± 0.141 | 20.9 ± 3.35 |

3.4. Total deposition of $^{137}$Cs

Total inventories of $^{137}$Cs in soil and standing biomass of Pinus kesiya, decay corrected to 1963, are summarized in Table 5. The sum of these inventories represents the total initial deposition up to 1963, assuming that losses via soil leaching and surface soil erosion have been negligible since then.
Table-5. Summarised estimates of initial deposition of $^{137}$Cs at Wat Chan (1963) in forest soil and aboveground biomass.

| Forest soil| Aboveground | Total |
|------------|-------------|-------|
| Tree internal| 21.8 | 669 |
| Needles | 4.2 | |

The calculated total deposition of $^{137}$Cs to pine forest at Wat Chan is consistent with previous studies in neighboring countries [8].

4. Conclusions

Approximately 50 years following deposition, $^{137}$Cs fallout from nuclear weapons tests is distributed at a very shallow depth in forest soil at Wat Chan, northern Thailand. Almost all of the $^{137}$Cs is distributed in the upper 5 cm of the soil profile with apparently negligible migration deeper into the soil. Calculated total deposition of $^{137}$Cs is consistent with previous studies and this could be representative of total deposition of $^{137}$Cs on nearby forests. Potassium appears not to be a good indicator of caesium migration in this forest system. However, stable caesium is a useful analogue when studying uptake of $^{137}$Cs by *Pinus kesiya* at Wat Chan, especially if the ‘available’ (NH$_4$-extractable) fraction of $^{137}$Cs within the soil is considered to be the primary source for uptake and recycling.

5. References

[1] Bunzl K and Kracke W 1988 Journal of Environmental Radioactivity 8 1-14
[2] IAEA 2010. Handbook of Parameter Values for the Prediction of Radionuclide Transfer in Terrestrial and Freshwater Environments, Vienna, International Atomic Energy Agency
[3] IAEA 2013 The International Nuclear and Radioecology Event Scale: Incident and Emergency Center
[4] Smith J and Beresford N A 2005 Chernobyl: Catastrophe and Consequences. Chichester, UK: Springer Praxis publishers.
[5] Cambray R S, Cawse P A, Garland J A, Gibson J A B, Johnson P, Lewis G N J, Newton D, Salmon L and Wade B O 1987 Observations on radioactivity from the Chernobyl accident. *Harwell Report AERE R* 12462 p 66
[6] Cambray R S, Playford K, Lewis G N J and Burton P J 1987 Radioactive fallout in air and rain: results for 1985 and 1986 *Harwell Report AERE R* 12872 p 26
[7] Hien P S, Binh P N, Truong N N, Bac V T and Ngo N T 1994 *J. Environ. Radioact.* 22 55–62
[8] Hien P D, Hiem H T, Quang N H, Huy N Q, Binh N T, Hai P S, Long N Q and Bac V T 2002 *J. Environ. Radioact.* 62 295 – 303
[9] Long N Q, Truong Y, Hien P D, Binh N T, Sieu L N, Giap T V and Phan N T 2012 *J. Environ. Radioact.* 111 53-8
[10] Wyttenbach A, Bajo S, Bucher J, Furrer V, Schleppi P and Tobler L 1995 *Z. Pflanzenernahr. Bodenw.* 158 499-504
[11] Varskog P, Neumann R and Steinnes E 1994 *J. Environ. Radioact.* 22 43-53
[12] Nongnuang S 2012 *Thai J. Fores*. 31 1-15