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FACULTY OF ENGINEERING  
DEPARTMENT OF ELECTRICAL AND COMPUTER ENGINEERING  
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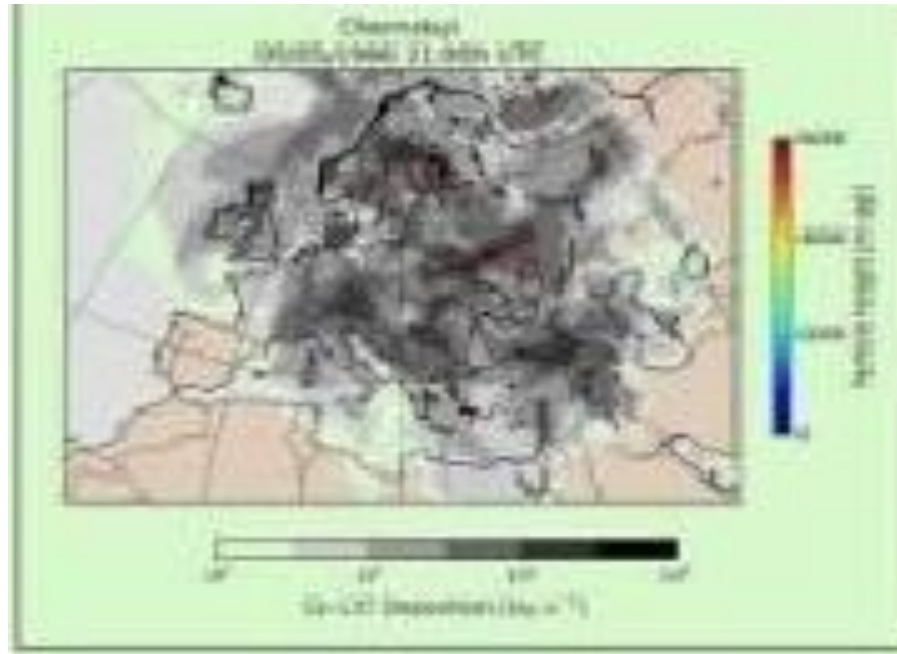
*A comparative analysis of Cs-137 soil migration over a thirty-six years study period (1987-2023) :*

*Experimental measurements vs compartment model predictions*

Alexandros Clouvas, Ioannis Kaissas, Stelios Xanthos

HNPS 2023

# Radioactivity spread out from Chernobyl in 1986



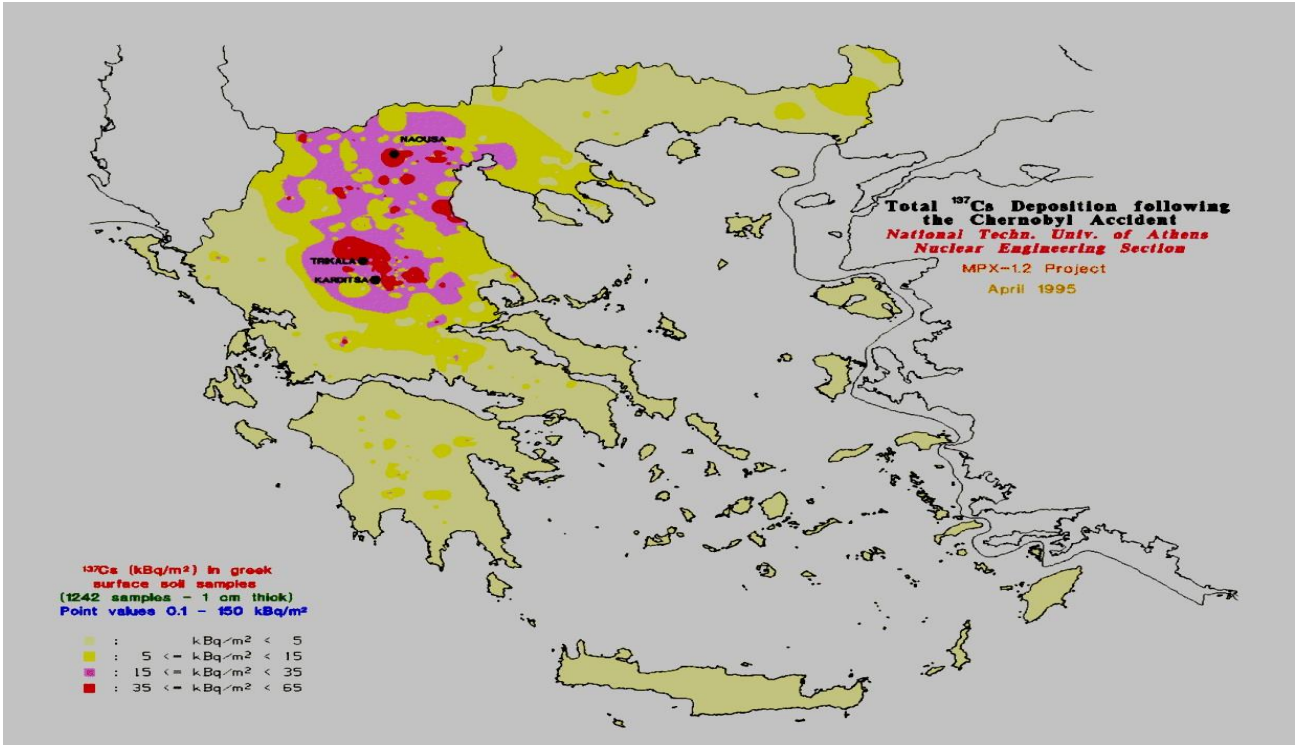
## Total Released Activities\*:

- **Cs-137**  $38 \cdot 10^{15}$  Bq
- **Cs-134**  $18 \cdot 10^{15}$  Bq
- **I-131**  $260 \cdot 10^{15}$  Bq
- **Sr-90**  $8 \cdot 10^{15}$  Bq
- **Pu-241**  $5 \cdot 10^{15}$  Bq

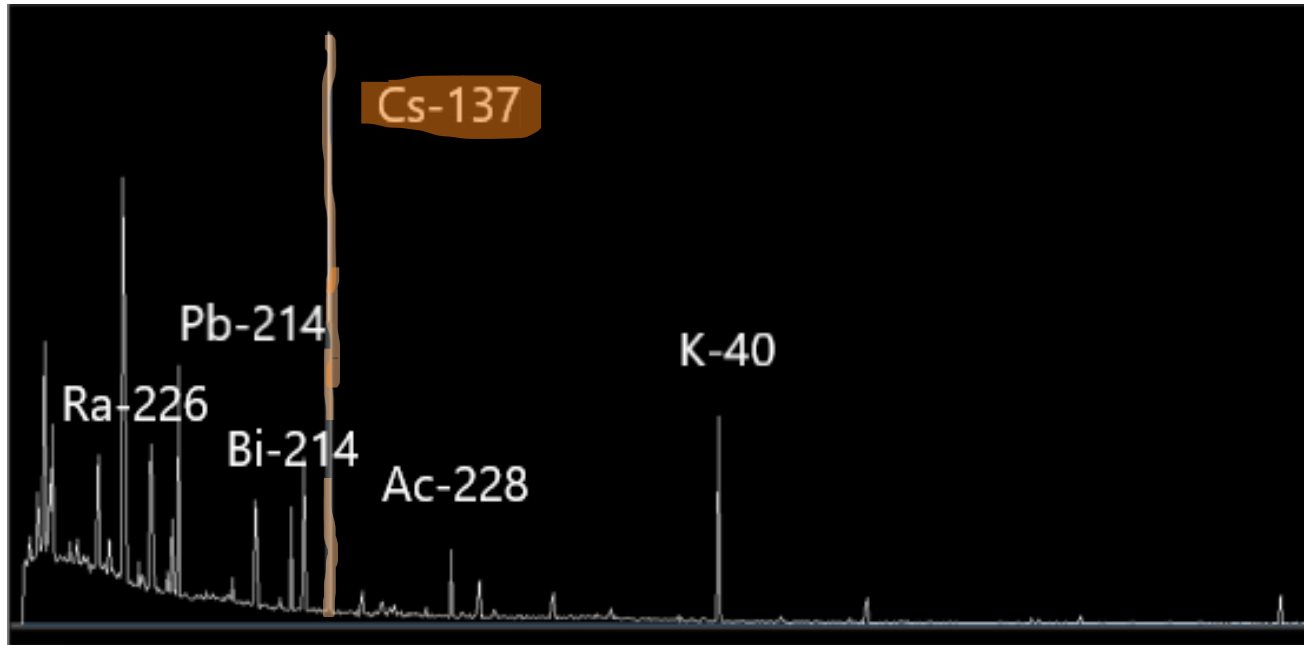
\*According to former Soviet Government

PetaBq

# Cs-137 deposition in Greece



# $\gamma$ -spectrum with High Purity Ge detector



- **Cs-137** with  $T_{1/2} = 30.2$  yrs
- **Cs-134** with  $T_{1/2} = 2.7$  yrs
- **I-131** with  $T_{1/2} = 8.0$  days

The **ratio** of the activities of **Cs-137** and **Cs-134** (backdated to May 1986) **in soil**, the first years after the accident when the latter was measurable, was found to **equals** approximately **2**.

Considering, :

- that the same ratio **2** was measured in air filters immediately after the arrival of the radioactive cloud in Greece, and
- that Cs-134 in soil, is only due to the Chernobyl accident (do not released by explosions of nuclear weapons),

we can assume that practically all Cs-137 in soil is due to the Chernobyl accident, **i.e. nuclear weapon tests fallout is negligible in Greece.**

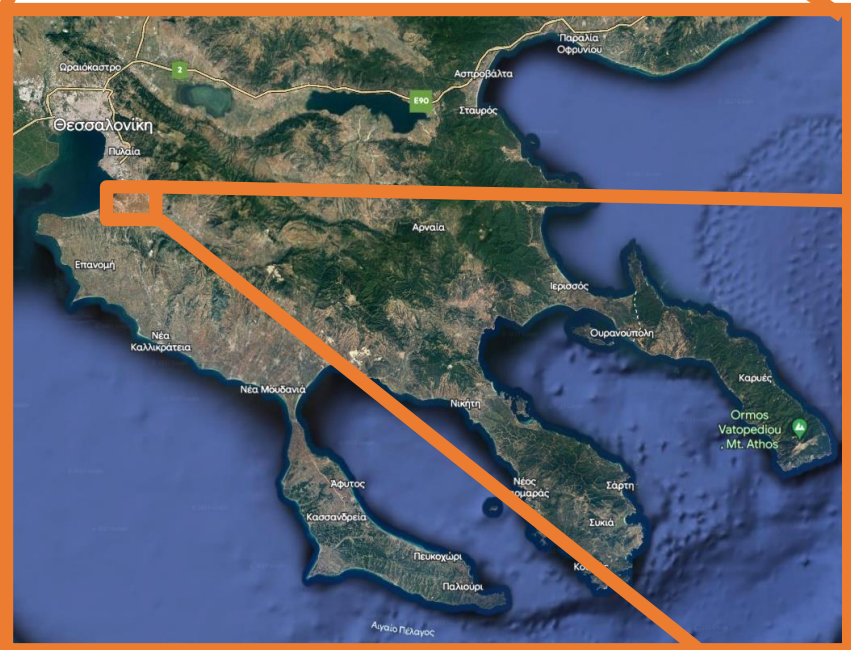
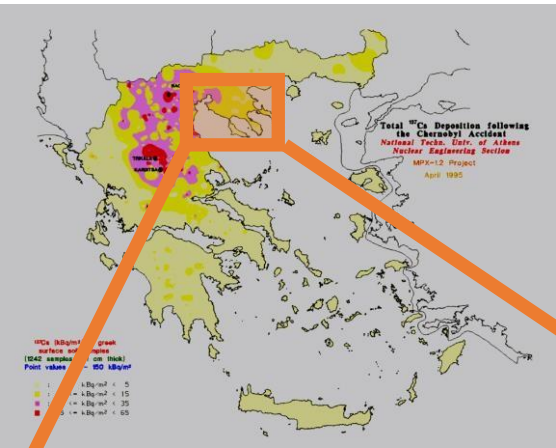


# Activity Concentration of Cs-137 vs Depth

The Nuclear Technology Laboratory of Aristotle University of Thessaloniki has conducted measurements on undisturbed soil located at the university farm, at the east-side of Thessaloniki.

On undisturbed soil, one can observe the kinematics of Cs-137, as it is moved slowly from the surface to the deeper layers of soil.

The non-disturbance of the particular area can be proven through the years due to the controlled agricultural activities in the university farm. No agricultural activities have been performed since 1986.





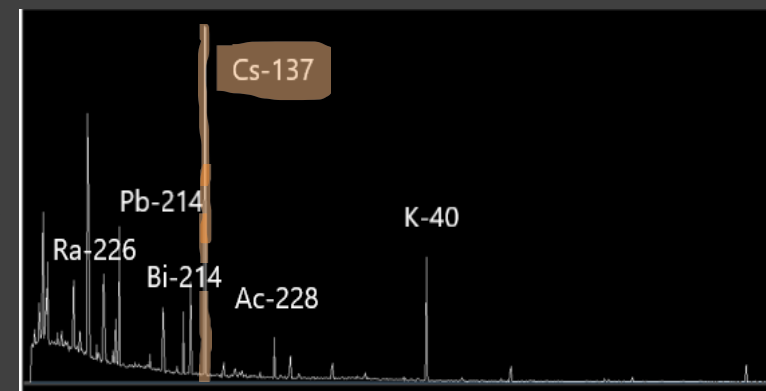
**In situ collection of soil**

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**Preparation of samples**

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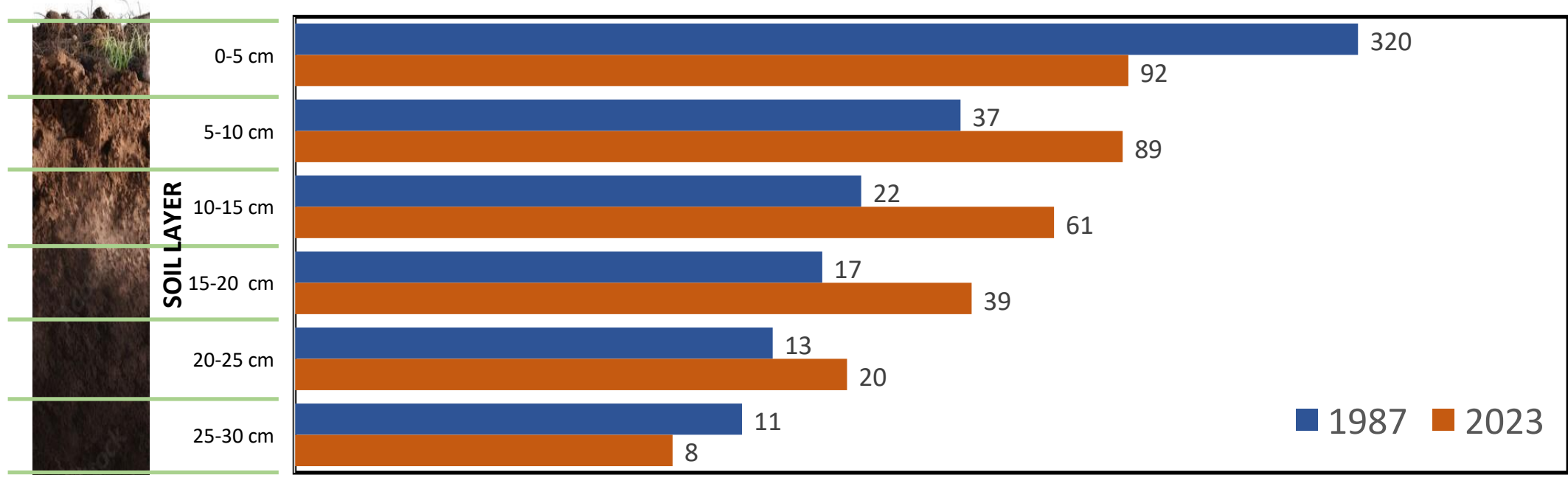
**Measurements in the HPGe**



# Measurement Results – Comparing 1987 and 2023

Cs-137 concentration *backdated for the time of Chernobyl accident* (Bq/kg)

1 10 100 1000



- Through the years Cs-137 migrates in deeper layers
- In the next years, the **maximum** concentration will **swift** from the **5 cm layer** to the **10 cm layer**
  - At deeper layers, the concentration ratio **R** increases through the years



# Compartment Model

$z$  : depth of soil layer (cm)

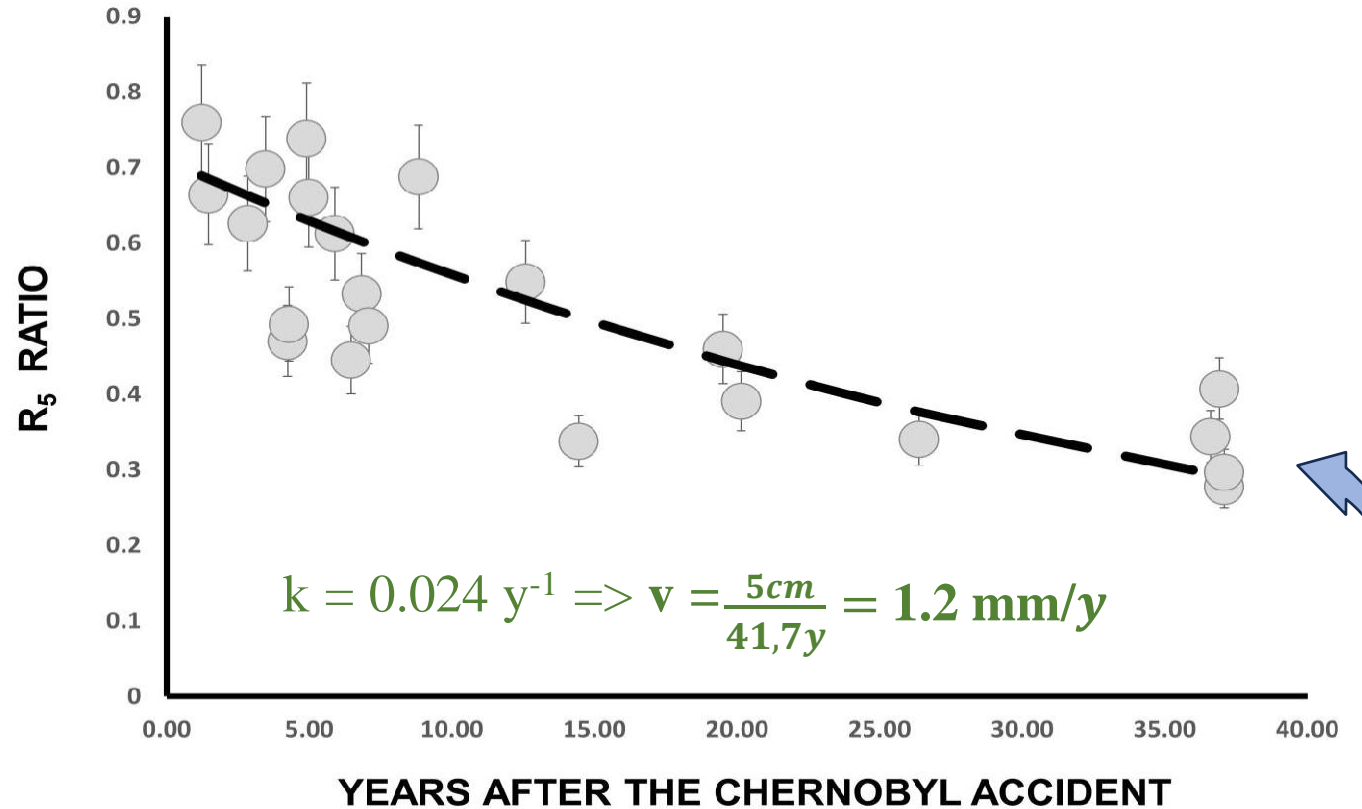
$T(z)$  : Cs-137 activity to units of surface and depth (Bq/m<sup>3</sup>)

$R_z$  : Ratio of the  $T(z)$  of a certain depth to the total  $T$  of all the layers

$$R_z = \frac{\int_0^z T(z) dz}{\int_0^{30} T(z) dz}$$

matching

Measurements



$$\frac{dR_5}{dt} = -kR_5$$



$$R_5(t) = R_5(t=0)e^{-kt}$$

$$\frac{dR_{10}}{dt} = kR_5 - kR_{10}$$



$$R_{10}(t) = [kR_5(t=0)t + R_{10}(t=0)]e^{-kt}$$

$$\frac{dR_{15}}{dt} = kR_{10} - kR_{15}$$



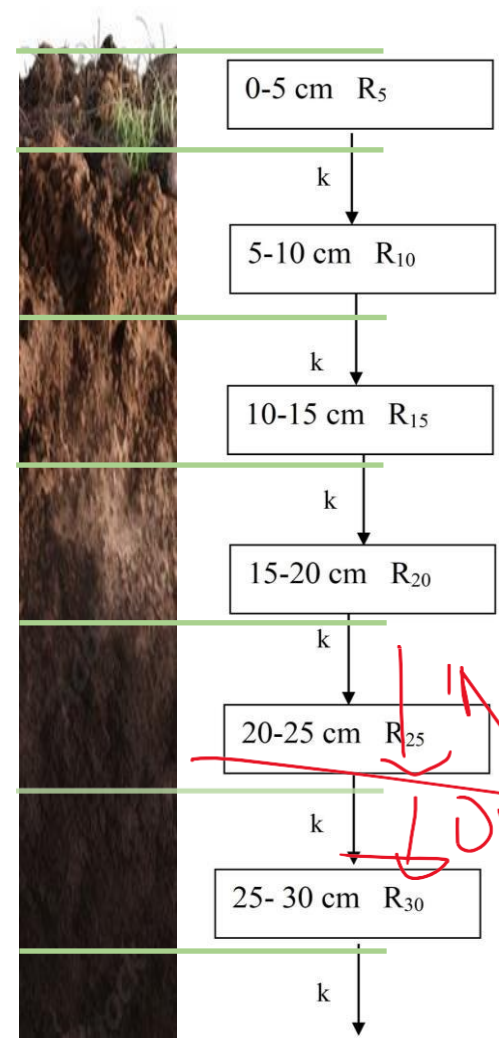
$$R_{15}(t) = \left[ \frac{k^2 R_5(t=0)t^2}{2} + kR_{10}(t=0)t + R_{15}(t=0) \right] e^{-kt}$$



$$\frac{dR_{20}}{dt} = kR_{15} - kR_{20} \implies R_{20}(t) = \left[ \frac{k^3 R_5(t=0)t^3}{6} + \frac{k^2 R_{10}(t=0)t^2}{2} + kR_{15}(t=0)t + R_{20}(t=0) \right] e^{-kt}$$

$$\frac{dR_{25}}{dt} = kR_{20} - kR_{25} \implies R_{25}(t) = \left[ \frac{k^4 R_5(t=0)t^4}{24} + \frac{k^3 R_{10}(t=0)t^3}{6} + \frac{k^2 R_{15}(t=0)t^2}{2} + kR_{20}(t=0)t + R_{25}(t=0) \right] e^{-kt}$$

$$\frac{dR_{30}}{dt} = kR_{25} - kR_{30} \implies R_{30}(t) = \left[ \frac{k^5 R_5(t=0)t^5}{120} + \frac{k^4 R_{10}(t=0)t^4}{24} + \frac{k^3 R_{15}(t=0)t^3}{6} + \frac{k^2 R_{20}(t=0)t^2}{2} + kR_{25}(t=0)t + R_{30}(t=0) \right] e^{-kt}$$



A simple **compartment model** used to simulate the distribution of Cs-137 in the soil through time. Each layer of soil is represented by a compartment.

Each differential equation considers the **Cs-137 input** from above and the **Cs-137 output** to the next deeper layer.

**k** stands for **transfer rate** between the compartments and its units are **years<sup>-1</sup>**

The simplicity lays on the use of **the same k for every different differential equation**.

It is acknowledged that **k** is not the same between consecutive soil layers and **increases with soil depth**, implying that **diffusion is the main mechanism of migration of Cs-137 ...**

... but in this way, the model can be **generalized well to new observations**, even if it does not excessively match the data.

INPUT k  
OUTPUT k

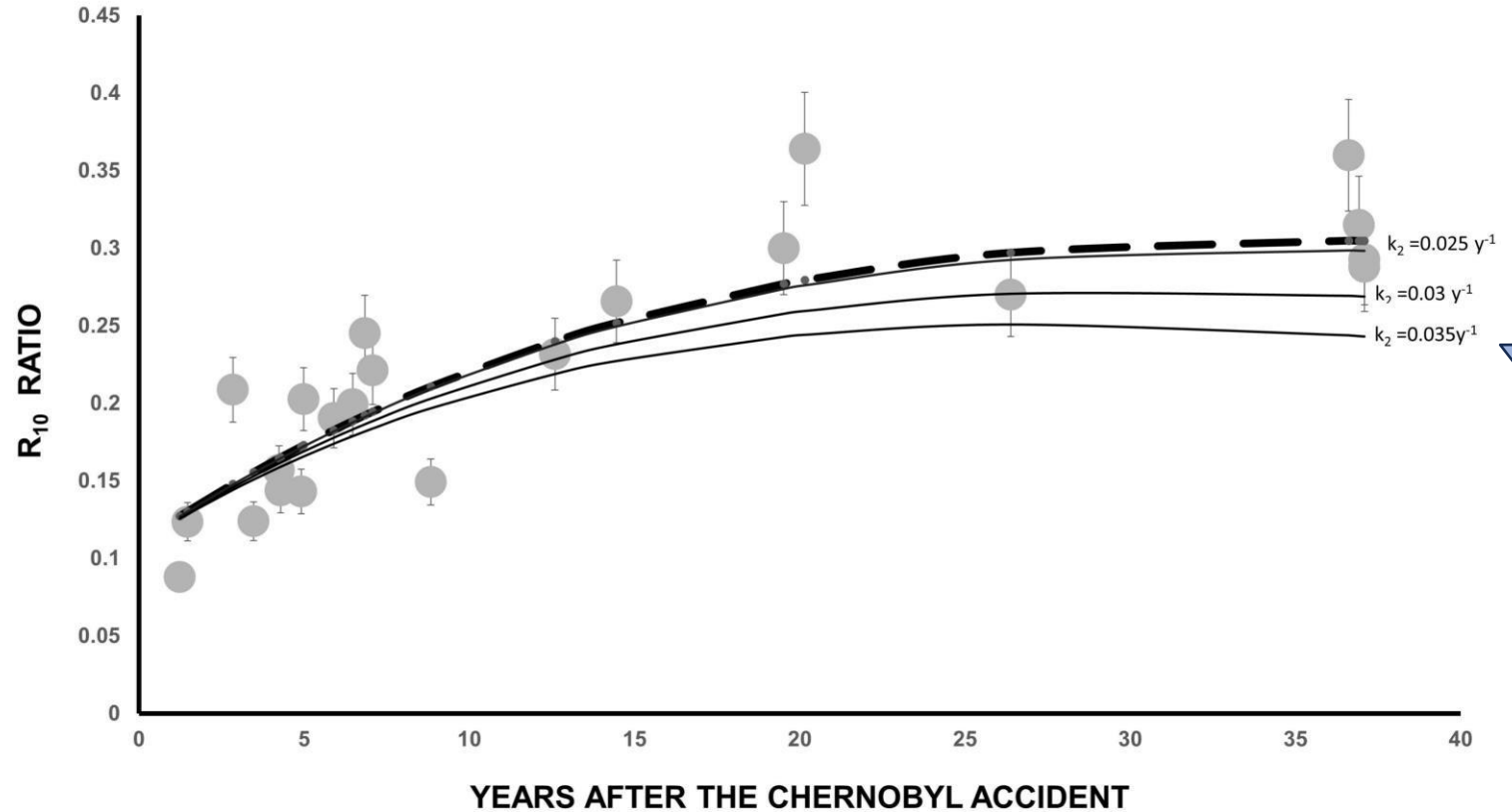
# Compartment model

Considering **diffusion** as the main mechanism of Cs-137 migration,  $k$  increases with depth (see Fick's law).

Such a model do not align with the observed outcomes.

This suggests that diffusion may not be the primary migration mechanism over long periods. Factors such as **advection** or other transfer processes may be more influential in the long-term migration of Cs-137.

with  $k_1, k_2, \dots, k_i$

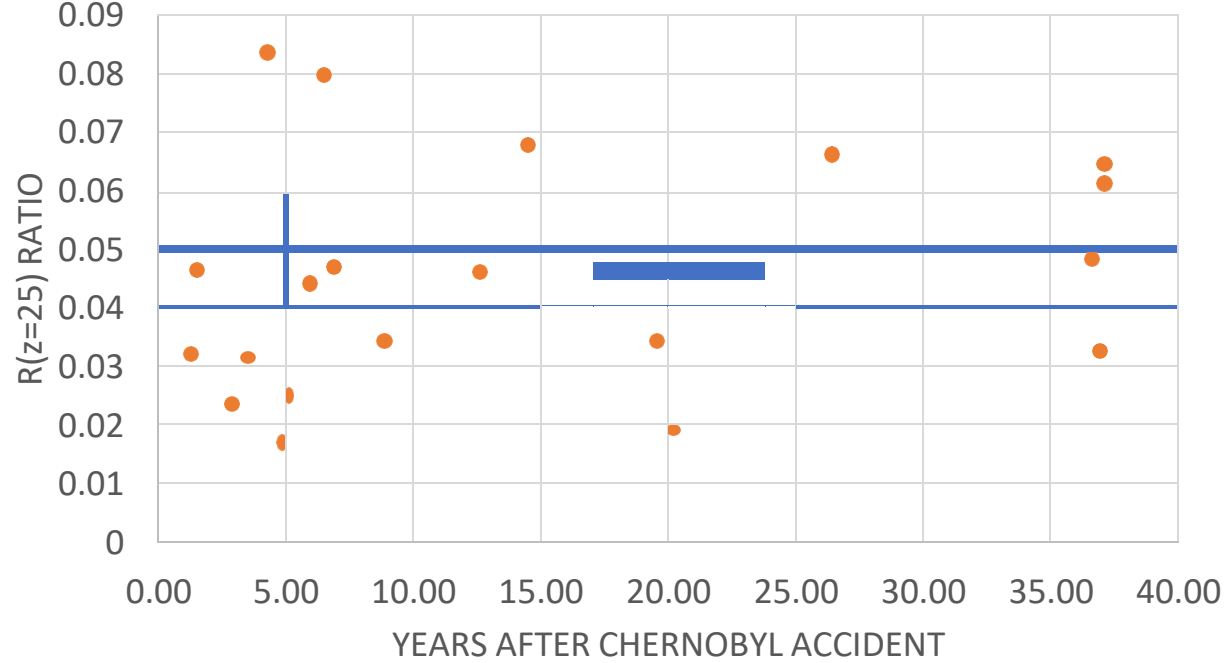
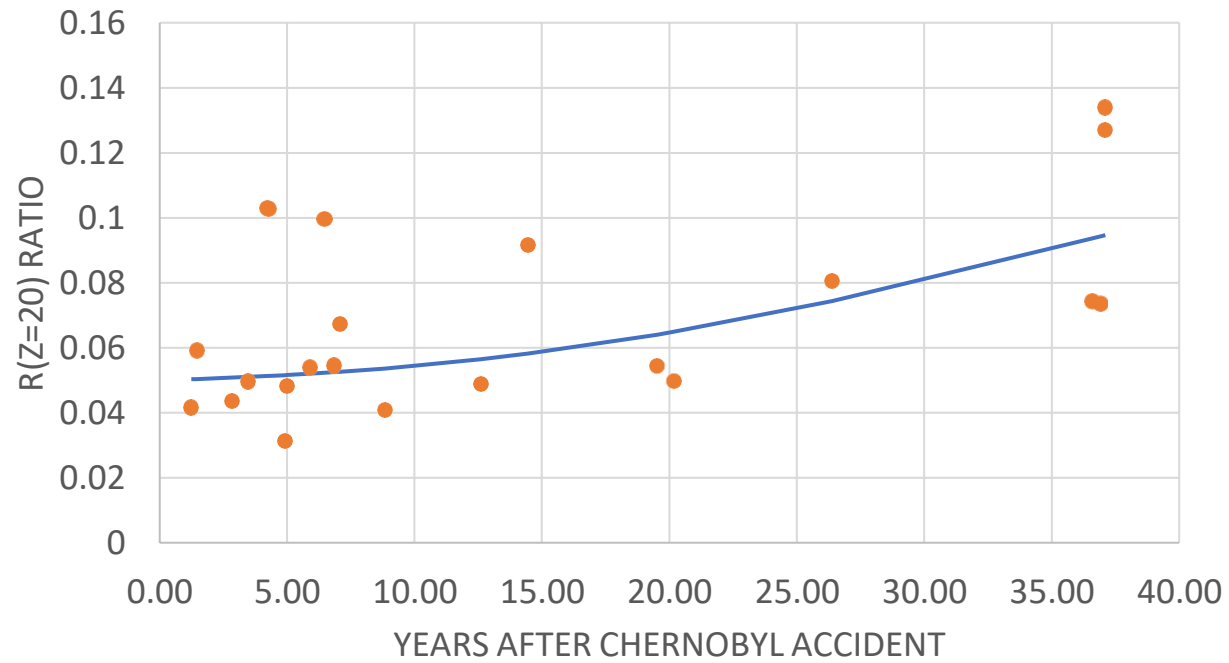
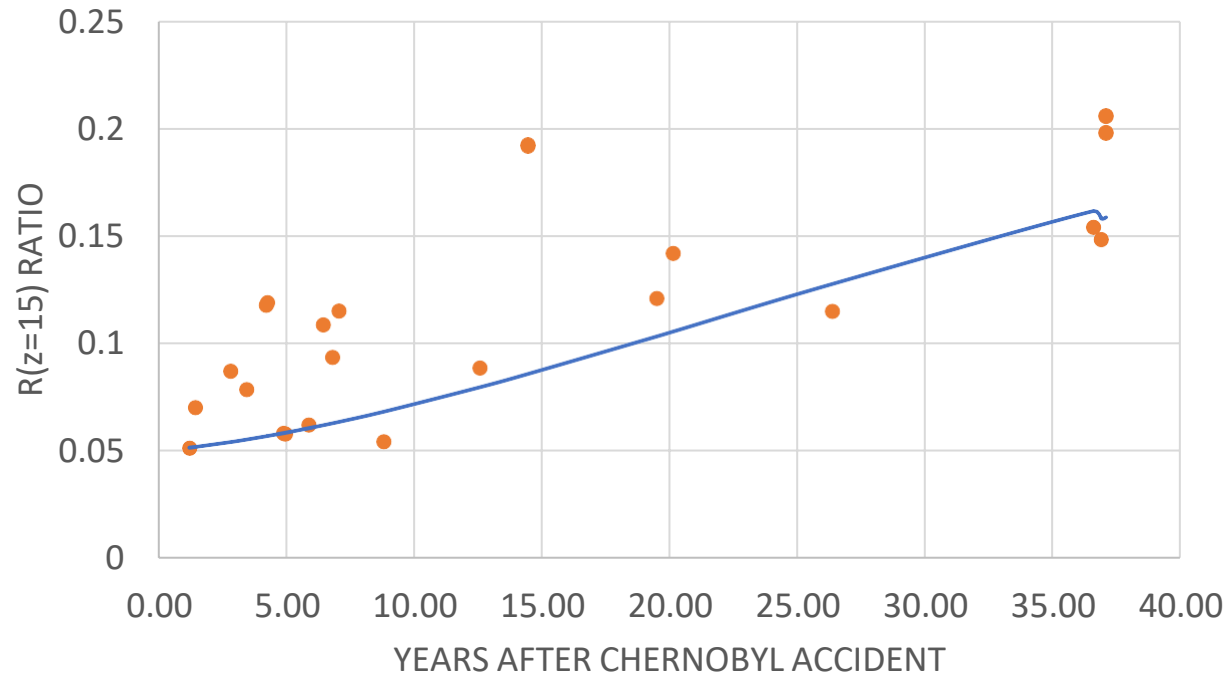
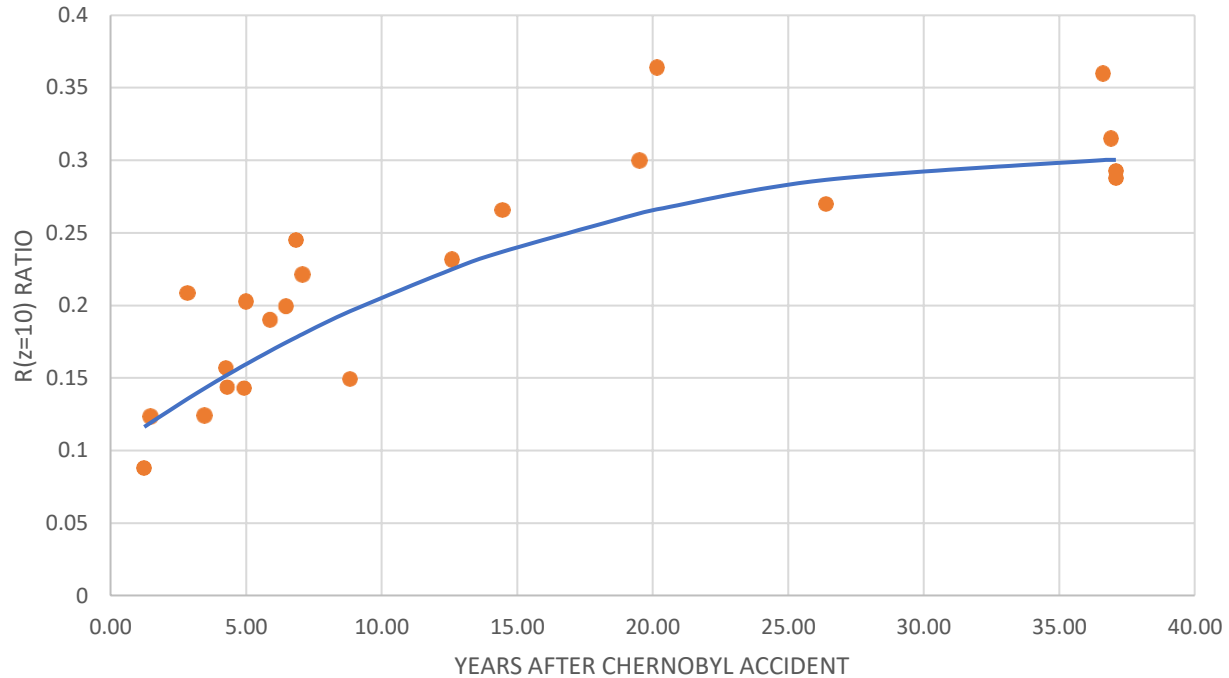


$$\frac{dR_5}{dt} = -k_1 R_5$$

$$\frac{dR_{10}}{dt} = k_1 R_5 - k_2 R_{10}$$

$$R_5(t) = R_5(t=0)e^{-k_1 t}$$

$$R_{10}(t) = \left( \frac{k_1 R_5(t=0)}{k_2 - k_1} \right) (e^{-k_1 t} - e^{-k_2 t}) + R_{10}(t=0) e^{-k_2 t}$$



## Deposition of Cs-137 (Bq/m<sup>2</sup>) in 2023 and 1986

$$A(t=36.6y) = 136 \frac{Bq}{Kg} * 1300 \frac{Kg}{m^3} * 0.05m = 8.8 \frac{kBq}{m^2}$$

Backdated to the time of Chernobyl accident in 1986:

$$A_0 = A(t = 36.6y) * 2^{\frac{36.6}{30.2}} = 20.2 \frac{kBq}{m^2}$$

This value matches with the total deposition at the area, which was independently measured to be about **27.3 kBq/m<sup>2</sup>** during the first year after the Chernobyl accident

# Just to Relax

Measuring U-238 and Th-232 series and K-40 yields the relevant concentrations for:

$$^{226}\text{Ra} = 13 \text{ Bq/kg}$$

$$^{228}\text{Ac} = 16 \text{ Bq/kg}$$

$$^{40}\text{K} = 228 \text{ Bq/kg}$$

Considering their uniform distribution on the soil and the factors of Lemercier studies for the conversion of activity to equivalent dose rate @ 1m above the ground:

$$\text{Equivalent dose rate for } ^{226}\text{Ra} = 7.33 \text{ nSv/h}$$

$$\text{Equivalent dose rate for } ^{232}\text{Th series} = 11.98 \text{ nSv/h}$$

Monte-Carlo simulations of the  $^{40}\text{K}$  distribution and the Cs-137 deposition yields:

$$\text{Equivalent dose rate @ 1m above the ground for } ^{40}\text{K} = 10.23 \text{ nSv/h}$$

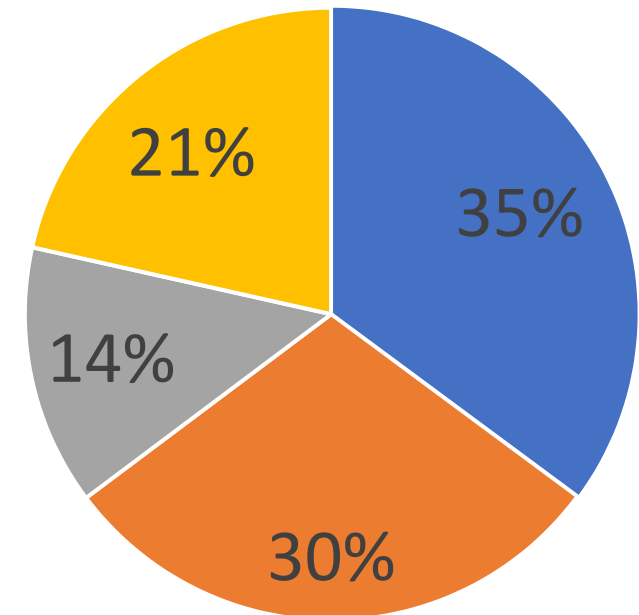
$$\text{Equivalent dose rate @ 1m above the ground for Cs-137} = 4.56 \text{ nSv/h}$$

$$\text{Total equivalent dose rate @ 1m above the ground} = 34.1 \text{ nSv/h.}$$

Measurements with handheld dosimeter on site reveals an Ambient Dose Equivalent Rate of about 40 to 45 nSv/h.

Contribution of Cs-137 and the naturally occurring radioisotopes to the Ambient Dose Equivalent Rate

■ Σειρά θορίου ■ K-40 ■ Cs-137 ■ Ra-226





# Just to Relax

Radiation Protection Dosimetry, 2023, 1–7

<https://doi.org/10.1093/rpd/ncad241>

Paper

OXFORD

## Long-term study (1987–2023) on the distribution of $^{137}\text{Cs}$ in soil following the Chernobyl nuclear accident: a comparison of temporal migration measurements and compartment model predictions

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### Abstract

After the Chernobyl accident, a designated area of  $\sim 1000\text{ m}^2$  within the University farm of Aristotle University of Thessaloniki in Northern Greece was utilized as a test ground for radioecological measurements. The profile of  $^{137}\text{Cs}$  in the soil was monitored from 1987 to 2023, with soil samples collected in 5-cm-thick slices (layers) down to a depth of 30 cm. The mean total deposition of  $^{137}\text{Cs}$  in the area, backdated to the time of the Chernobyl accident, was determined to be  $18.6 \pm 1.8\text{ kBq m}^{-2}$  based on four follow-up profile measurements of  $^{137}\text{Cs}$  in the soil for the years 2022 and 2023. It is noteworthy that this value is similar to the total deposition at the site, which was independently measured to be about  $20\text{ kBq m}^{-2}$  during the first year after the Chernobyl accident. The fractional contribution of each soil layer (e.g., 0–5 cm, 5–10 cm, 10–15 cm, etc.) to the total deposition of  $^{137}\text{Cs}$  (0–30 cm) is presented and analyzed. A compartment model was utilized to forecast the temporal evolution of fractional contributions of the different soil layers to the total deposition of  $^{137}\text{Cs}$  (0–30 cm). In this model, each soil layer is represented as a separate compartment. The model assumes that the transfer rates between adjacent compartments are equal. The agreement between the measured fractional contributions and the model predictions suggests that the compartment model with equal transfer rates can capture the broad patterns of  $^{137}\text{Cs}$  migration within the soil layers over the long period of 1987–2023. However, the use of a second compartment model with increasing transfer rates between consecutive soil layers did not align with the observed outcomes. This indicates that diffusion may not be the primary migration mechanism over the 36-y period covered by our study.

### Introduction


The long-term external dose resulting from nuclear accidents, such as Chernobyl and Fukushima, primarily arises from the deposition of  $^{137}\text{Cs}$  in the environment. Accurate knowledge of the distribution of  $^{137}\text{Cs}$  deposited in the soil is crucial for reliable assessments of the external dose<sup>(1, 2)</sup> and for understanding the potential uptake of this radionuclide by plants through root systems. Vertical migration of  $^{137}\text{Cs}$  in the soil has been a subject of extensive research<sup>(3, 4)</sup> because of its importance in assessing environmental impacts and potential risks. The radioecological studies conducted following the Chernobyl accident have provided valuable insights and experience that could aid in the planning and interpretation of similar studies in

Japan following the Fukushima accident. By leveraging the knowledge gained from the Chernobyl studies, researchers could enhance the effectiveness and efficiency of radioecological investigations in Fukushima.

After the Chernobyl accident, a designated area of  $\sim 1000\text{ m}^2$  within the University farm of Aristotle University of Thessaloniki in Northern Greece was utilized by the Nuclear Technology Laboratory as a test ground for radioecological measurements<sup>(5, 6, 7)</sup>. In the present study, follow-up profile measurements of  $^{137}\text{Cs}$  in the soil were conducted for the years 2022 and 2023 within the designated area. The measurements of the present study were performed  $\sim 10\text{ y}$  after the last measurements<sup>(7)</sup> taken at the site, which spanned from 1987 to 2012. To analyze the data, the time evolution

Received: June 28, 2023. Revised: July 31, 2023. Editorial decision: August 7, 2023. Accepted: August 15, 2023

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