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Experimental and Modelling Studies of Radionuclide Migration from Contaminated Groundwaters

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Abstract
Lysimeter-based studies of radionuclide uptake by winter wheat are being undertaken to investigate soil-to-plant transfer processes. A five year multi-disciplinary research project has concentrated on the upward migration of contaminants from near surface water-tables and their subsequent uptake by a winter wheat crop. A weighted transfer factor approach and a physically based modelling methodology, for the simulation and prediction of radionuclide uptake, have been developed which offer alternatives to the traditional transfer factor approach. Integrated hydrological and solute transport models are used to simulate contaminant movement and subsequent root uptake. This approach enables prediction of radionuclide transport for a wide range of soil, plant and radionuclide types. This paper presents simulated results of ^{22}Na plant uptake and soil activity profiles, which are verified with respect to lysimeter data. The results demonstrate that a simple modelling approach can describe the variability in radioactivity in both the harvested crop and the soil profile, without recourse to a large number of empirical parameters. The proposed modelling technique should be readily applicable to a range of scales and conditions, since it embodies an understanding of the underlying physical processes of the system. This work constitutes part of an ongoing research programme being undertaken by UK Nirex Ltd., to assess the long term safety of a deep level repository for low and intermediate level nuclear waste.

Introduction
While a large international database exists on the topic of soil-to-plant transfer of radionuclides from soils with contaminated surface horizons, the question of crop uptake of radioactivity from deeper contaminated regions within soil profiles remains virtually unexplored. Of particular interest is the situation which may arise in connection with the disposal of radioactive wastes to shallow or deep repositories where groundwater movement is likely to be the principal vector of radionuclide emergence into the rooting region of plants. The main questions surrounding this scenario are i) the likely magnitude of up-profile transport of both sorbed and conservative radionuclides as a result of physical and biological processes, and ii) the quantification of soil-to-plant transfer in a system in which the majority

of radioactivity lies below the depth at which the bulk of plant roots occur. Under these circumstances the classical transfer factor approach cannot be applied directly and amended transfer factor methods or alternative modelling techniques, which incorporate underlying physical processes, must be sought. These problems are currently being addressed in an ongoing lysimeter study and augmentative modelling programme being undertaken at Imperial College, UK, and some results of both the experimental and modelling work are presented here.

Lysimeter System Design

The Imperial College experimental lysimeter facility was built in the early 1960s and consists of 28 lysimeters in two rows of 14. In 1988, eight of these lysimeters were refurbished and filled with two depths of a sandy loam soil (Eutric Cambisol), the four "shallow" lysimeters have a soil depth of 40cm whilst the four "deep" have a depth of 70cm. Beneath the soil is a 20cm inert substrate of polyethylene beads and these are separated by a thin geotextile layer. All the lysimeters have a control system which maintains the water table at a height of 5cm above the geotextile layer with an accuracy of 0.5cm (Figure 1). The control system uses optical switches to control peristaltic pumps which import or export water from the lysimeter to a common reservoir which is dosed annually with carrier-free sources of radionuclides. Any evapotranspiration from the lysimeter will result in the net import of contaminated water to the water table; and further movement of individual radionuclides upwards from the water table will be controlled by physicochemical solute transport mechanisms as well as biological translocation.

Cropping Regime and Analysis

Six radionuclides were introduced into the experimental system in 1990 (^{137}Cs,^{60}Co,^{109}Cd,^{22}Na,^{36}Cl and ^{99}Tc) and these have been "topped up" annually at intervals timed to coincide with the beginning of the summer growth period. The lysimeters are cropped with winter wheat (Triticum aestivum); this is planted in November/December and harvested in July/August the following year. During its growth period the crop's performance is monitored by a series of shoot growth measurements (including crop height and leaf area index) and root density determinations made *in situ* using "Perspex" access tubes into which a rigid fiberoptic endoscope ("Borescope") may be inserted. After harvest, radionuclide determinations are made in shoot, leaf, chaff and grain tissues. These measurements are complemented with soil core analyses in which the vertical distributions of individual radionuclides are determined. As well as plant and soil measurements, radiochemical activities within the lysimeter system are recorded on a fortnightly basis.

Hydrological and Meteorological Measurements

An automatic weather station adjacent to the site provides detailed climatological data, logged on a 1/100th of a day timestep (approx 15 mins.). These include solar and net radiation, rainfall, air temperature, humidity, and wind speed and direction. A series of tensiometers and thermistors provide soil water pressure and temperature measurements, also at a 1/100 day timestep, at 10cm intervals down the soil profile. The soil water content distribution is obtained on a weekly basis using a time domain reflectometer (TDR) and a neutron probe. In addition, the fluxes into and out of the lysimeters (as recorded by the number of revolutions of the peristaltic pumps) enable the water balance within the lysimeters to be accurately monitored.

Weighted Soil-to-Plant Transfer Factors

For the purposes of radiological dose prediction quantitative estimates are required of radionuclide activity concentrations within plant tissues at any stage in the plant's ontogeny when it may be consumed by an animal or human. Experimentally, transfer coefficients provide a useful measure of the overall efficiency of contaminant movement from donor to receptor. However, the International Union of Radioecologists stipulates that soil-to-plant transfer factors should be "based on dry plant matter and oven dry soil, and the assumption that all radioactivity is present in the soil layer of 0-10cm for grass and of 0-20cm for all other crops" (IUR, 1989). In the present study this definition is clearly untenable, as the bulk of the activity lies at a depth below 20cm. For this reason the concept of a "weighted transfer factor" (TF_w) has been adopted, which takes into account the accessibility of radionuclides, in a given layer, to the plant. It assumes that this accessibility is directly related to the degree of exploitation by the plant roots, since the denser the root system in any layer the greater the probability of absorption. The dimensionless "weighted transfer factor" is calculated thus:

$$TF_w = \frac{[R]_{plant}}{\sum_{i=1}^{n} ([R]_i f_i)} \qquad (1)$$

where $[R]_{plant}$ is the specific radionuclide activity in the plant, $[R]_i$ is the specific radionuclide activity in the i^{th} soil layer and f_i is a weighting factor defined as:

$$f_i = \frac{abundance\ of\ roots\ within\ i^{th}\ soil\ layer}{abundance\ of\ roots\ within\ the\ entire\ soil\ profile} \qquad (2)$$

This is obtained using direct measurements of root density, taken with the Borescope.

The Use of Combined Hydrological and Solute Transport Models to Simulate Contaminant Migration

The use of transfer factors provides a comparative measure of the behaviour of radionuclides within the soil plant system, without inferring any mechanistic information. However this lack of physical process representation has drawbacks, transfer factors only refer to instantaneous "snapshots" of the distribution of contamination in the system at the time of sampling and it is therefore difficult to use single soil-to-plant transfer factors to infer dynamic radionuclide movement. An alternative to the transfer factor approach is the use of dynamic models such as the combined hydrological and solute transport models described by Butler and Wheater (1990). These models are based on mechanistic principles of unsaturated flow processes and physical solute transport, coupled with a quasi-mechanistic plant absorption representation.

The use of this type of modelling approach, which considers the underlying physical processes of the system, means that the hydrological fluxes, which are the primary driving forces behind radionuclide movement, are incorporated.

The Soil-Plant-Water Model

The hydrological model enables simulations of the lysimeter soil water hydrology to be undertaken. Time dependant soil water movement is represented through a one-dimensional form of Richards' equation which incorporates evapotranspirative loses (U_w):

$$\frac{d\theta}{dt} = \frac{\partial\left[k(\psi,z)\left[\frac{\partial\psi}{\partial z}+1\right]\right]}{\partial z} - U_w \qquad \textbf{(3)}$$

This equation relates the change of soil moisture content θ to the hydraulic flux divergence at an elevation z and incorporates Darcy's law which assumes that the flux is linearly related to the total hydraulic head gradient. The relationships of soil moisture content and hydraulic conductivity (k) with matric potential (ψ) for the lysimeter soil are represented using parametric functions derived from experimental data (Butler and Wheater, 1990).

The Solute Transport Model

The contaminant transport model is based on an advection-dispersion approach, which utilises the soil water contents and fluxes obtained from the hydrological model. The rate of change of contaminant stored within a unit volume is related to the divergence of an advective flux due to bulk water motion and a dispersive flux resulting from the combination of molecular diffusion and mechanical dispersion. The advective flux is given by qc where q is the Darcy water flux and c is the contaminant concentration. Dispersion arises from the movement of the contaminant through the tortuous microscopic pathways within the soil pore spaces. The dispersive flux is assumed to be linearly related to the vertical concentration gradient through the hydrodynamic dispersion parameter D_h. The storage of contaminant in the soil is partitioned between the solute and sorbed phases. The activity of contaminant stored in solution per unit volume is given as θc, whilst the activity of sorbed contaminant per unit soil mass is given as S. The basic model equation, taking into account losses due to root uptake (U_s) and radioactive decay (Γ), takes the form:

$$\frac{\partial[\theta c+S]}{\partial t}=-\frac{\partial\left[\theta D_h\frac{\partial c}{\partial z}-qc\right]}{\partial z}-U_s-\Gamma \qquad \textbf{(4)}$$

Sorption of contaminant onto the surface of the soil matrix (of dry bulk density ρ_b) is assumed to be represented by a linear partition coefficient (K_d), hence:

$$S=\rho_b K_d c \qquad \textbf{(5)}$$

The uptake of solute by the plant root system is represented using a linear form of Michaelis-Menton rate kinetics (Epstein, 1966; Nye and Tinker, 1977; Barber, 1984). It is underlain by the assumptions that the concentrations at the root boundary are small, that uptake efficiencies for any section of the entire root system are identical, and that the concentration at the root/soil boundary is equal to the mean soil water concentration of the layer. If α is the root sorption coefficient, a is the average root radius and ρ_r is the root density, obtained through "Borescope" readings, root uptake is given as:

$$U_s=2\pi a\alpha\rho_r c \qquad \textbf{(6)}$$

The loss due to radioactivity (Γ) is represented through the use of the decay constant λ, hence:

$$\Gamma=\lambda(\theta c+S)=\lambda(\theta+\rho_b K_d)c \qquad \textbf{(7)}$$

Results and Discussion

Model simulations for the 1990, 1991 and 1992 crop harvests and the corresponding TF_ws derived from the observed data, are shown for ^{22}Na, using a Julian Date system, which has the form *YYddd* (where *YY* is the last 2 digits of the year and *ddd* is the cumulative day number, the Julian day).

The observed mean values of plant uptake at harvest, compared with model simulation predictions, are given in table 1 and figure 2. The average distributions of soil specific activities obtained from the model simulations are presented in figures 3 to 5.

Figure 2 shows the average observed and simulated crop uptake for each harvest and table 1 shows, in addition, the associated standard errors. It can be seen that the simulated values give a reasonable replication of the harvested activities for both deep and shallow lysimeters, and that the simulated values generally lie within the limits of the experimental data.

Figure 3a shows a good match between the observed and simulated shallow lysimeter soil activity profile for the first crop harvest (JD 90207, i.e. 26th July 1990) and although figure 3b shows an overestimation for the deep lysimeter activity, the general trend is replicated. In figures 4a and 4b the simulations of soil profile activities for the second harvest (JD 91226, i.e. 14th August 1991) show excellent agrement for both deep and shallow lysimeters. In figures 5a and 5b the results for the third harvest (JD 92209, i.e. 29th July 1992) show a slight under estimation of the activities but good replication of the overall profile shape. It is interesting to note that the lysimeter activities for 1990 were considerably larger than those for 1991 and 1992, this is due to the high evaporation rates present during 1990 which resulted in significant radionuclide migration up the profile.

The weighted transfer factors for each of the plant components are shown in figure 6, with 6a, b, c and d referring to the grain, chaff, leaf and stem respectively. The results show a highly significant year-on-year change in TF_w but this is not systematic. They also show that the TF_w values derived for "deep" lysimeters were considerably higher than those for "shallow" lysimeters. This may suggest that the efficiency of contaminant uptake is greater in the deeper soil layers. This could be due to the greater hydraulic conductivity in these deeper, and hence wetter, regions which ensures a more efficient supply of ions to the root surface. This concept has not been considered in the solute transport model simulations, and it may account for the underestimation of simulated uptake in the deep lysimeters for 1991 and 1992.

Conclusions

A lysimeter experiment has been designed to investigate the vertical migration of radionuclides through soil profiles from contaminated groundwaters. The experimental design is novel because it considers upward movement of radionuclides, rather than the more conventional surface deposition scenario. The whole process has been assessed in two ways: through the use of weighted transfer factors; and through a linked hydrological and solute transport model. The model is driven by observed hydrological stresses and a knowledge of soil and plant properties, and is able to reproduce the time varying soil profile and crop uptake activity values. The results obtained for the simulation of ^{22}Na are very encouraging, especially if it is considered that the model parameters were calibrated for 1990 and 1991 and applied in a predictive manner to data for 1992. The results presented herein are for ^{22}Na but the modelling scheme is currently being applied to other radionuclides. However, there is still some uncertainty in the conceptualisation of plant uptake, and in this respect the weighted transfer factor results have highlighted an area for further model development.

The combined hydrological and solute transport models presented here are able to provide accurate predictions without recourse to a large number of parameters. It is hoped that the portability of the modelling methodology will mean that parameters derived at the plot scale can be applied to field scale scenarios. A series of column experiments are currently being undertaken to establish the parameter values for a range of plants, soil types and radionuclide species and their relationship to those identified from the lysimeter experiments.

Acknowledgements
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Table 1: Simulated and observed harvest crop activities (MBq per m^2)

Year	Lysimeter (generic)	Simulated	Observed	
		mean	mean	standard dev.
1990	Deep	0.28	0.32	0.19
1990	Shallow	0.69	0.91	0.53
1991	Deep	0.02	0.19	0.11
1991	Shallow	0.77	0.79	0.28
1992	Deep	0.01	0.16	0.13
1992	Shallow	0.72	1.11	0.51

Figure 1: Schematic cross-section of a lysimeter showing the water table control system.

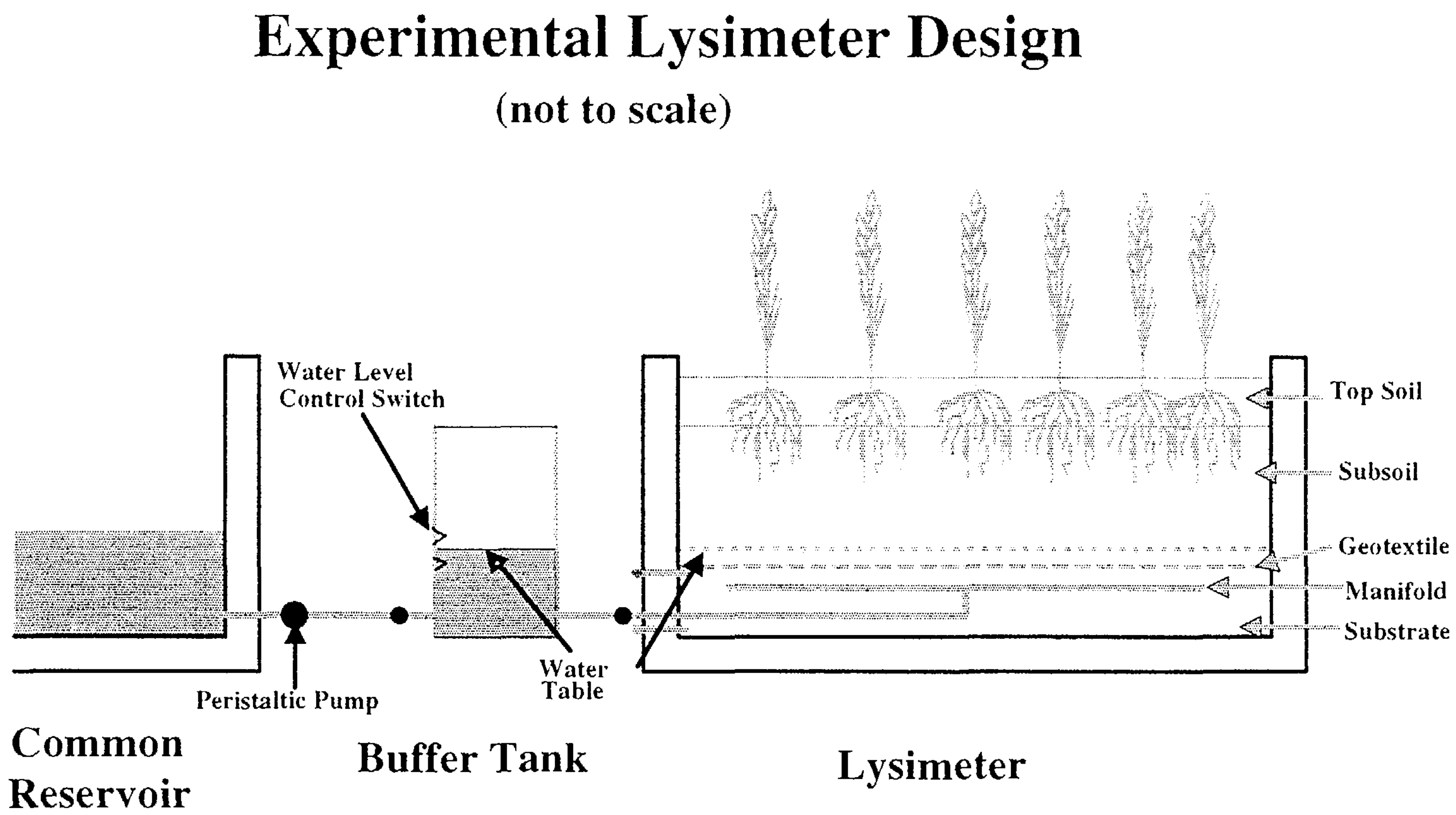

Figure 2: Simulated and observed harvest crop activities (MBq per m^2)

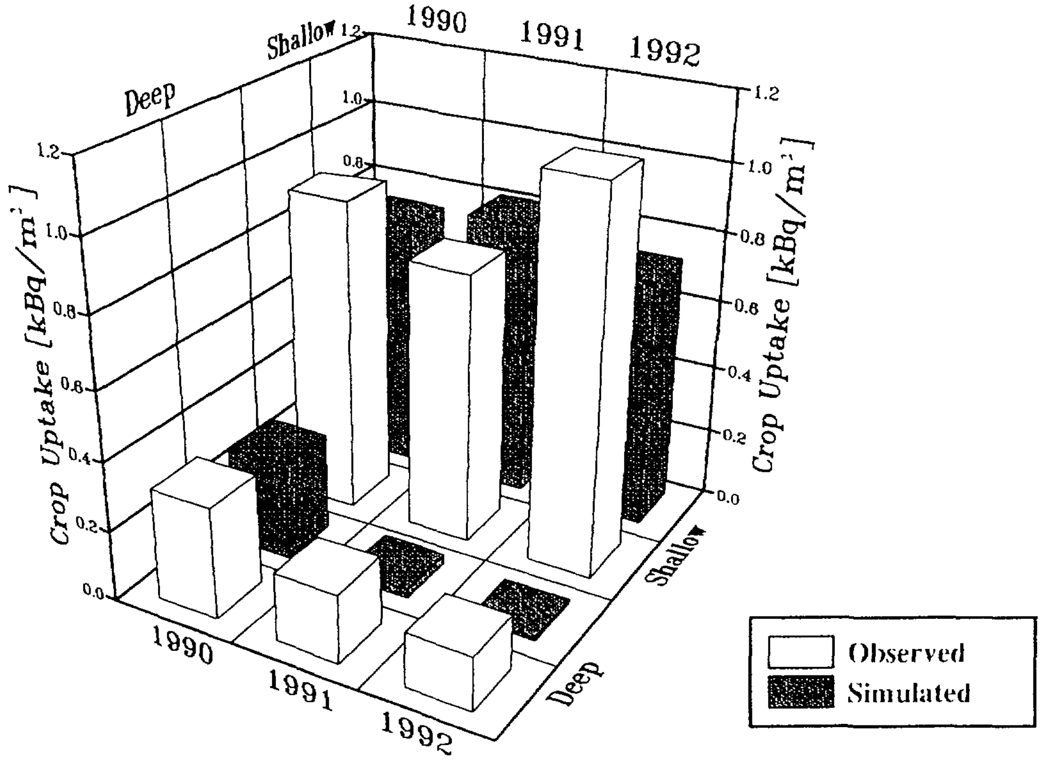

Figure 3a: Shallow lysimeter specific activity depth profile Julian Date 90207

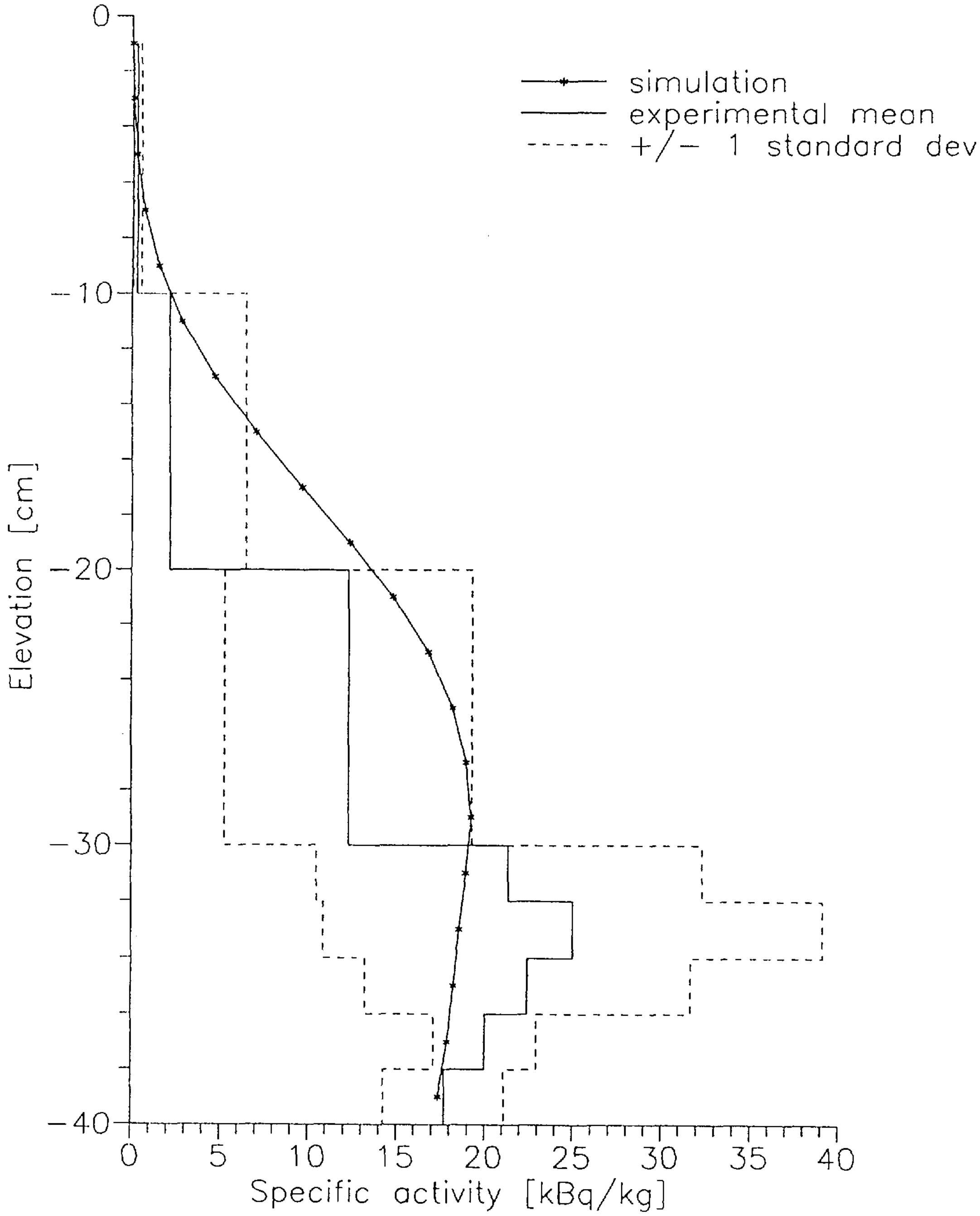

Figure 3b: Deep lysimeter specific activity depth profile Julian Date 90207

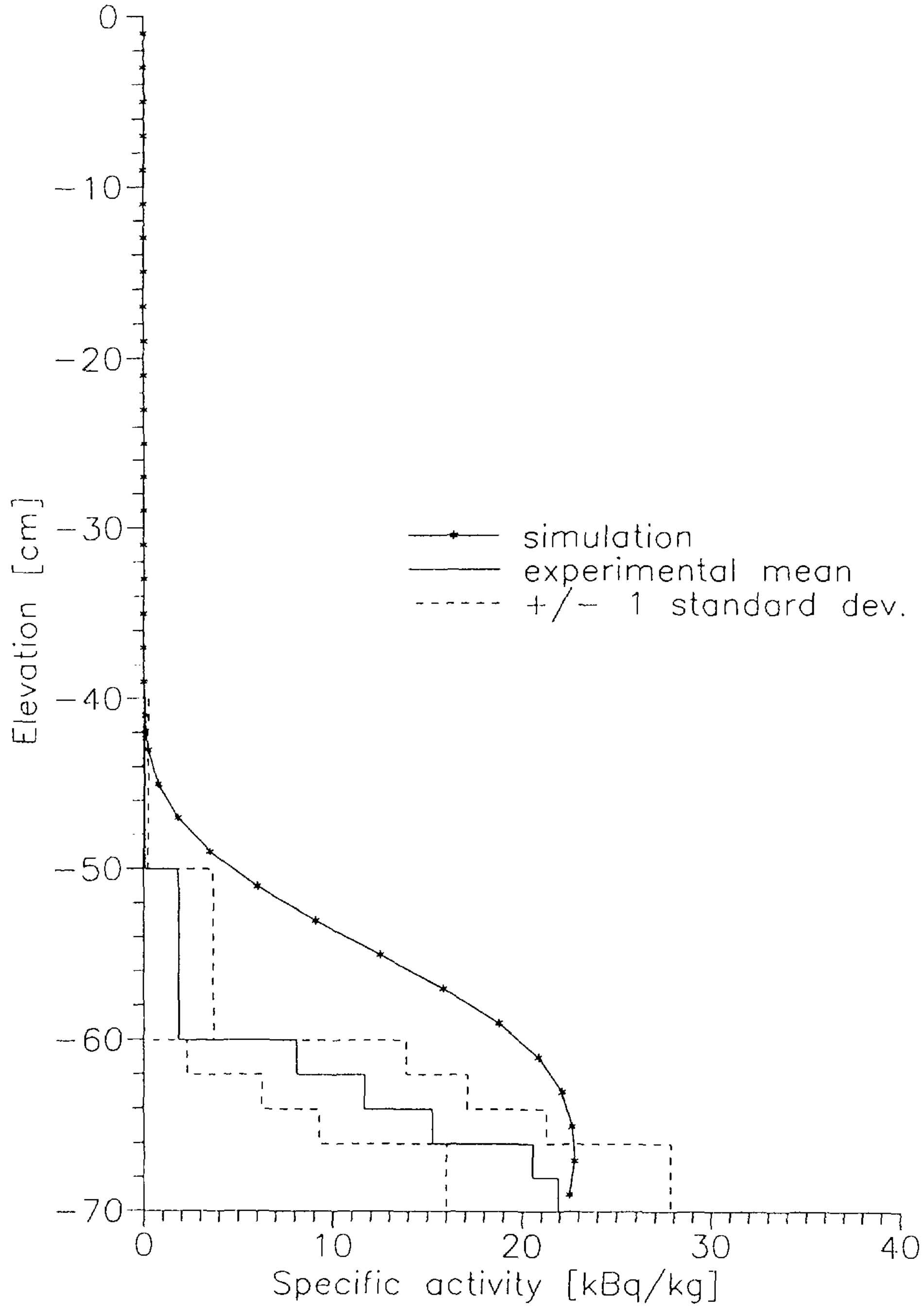

Figure 4a: Shallow lysimeter specific activity depth profile Julian Date 91226

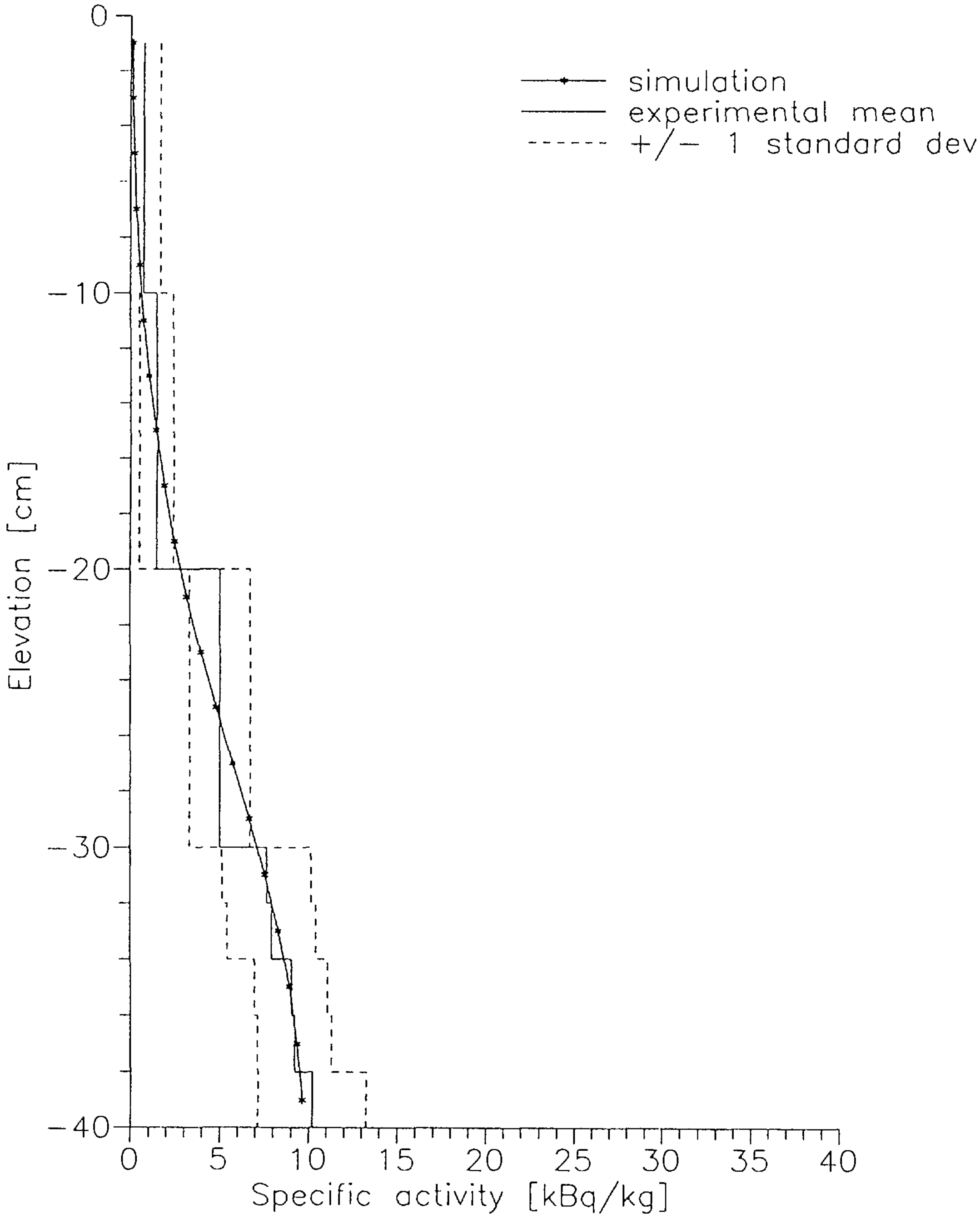

Figure 4b: Deep lysimeter specific activity depth profile Julian Date 91226

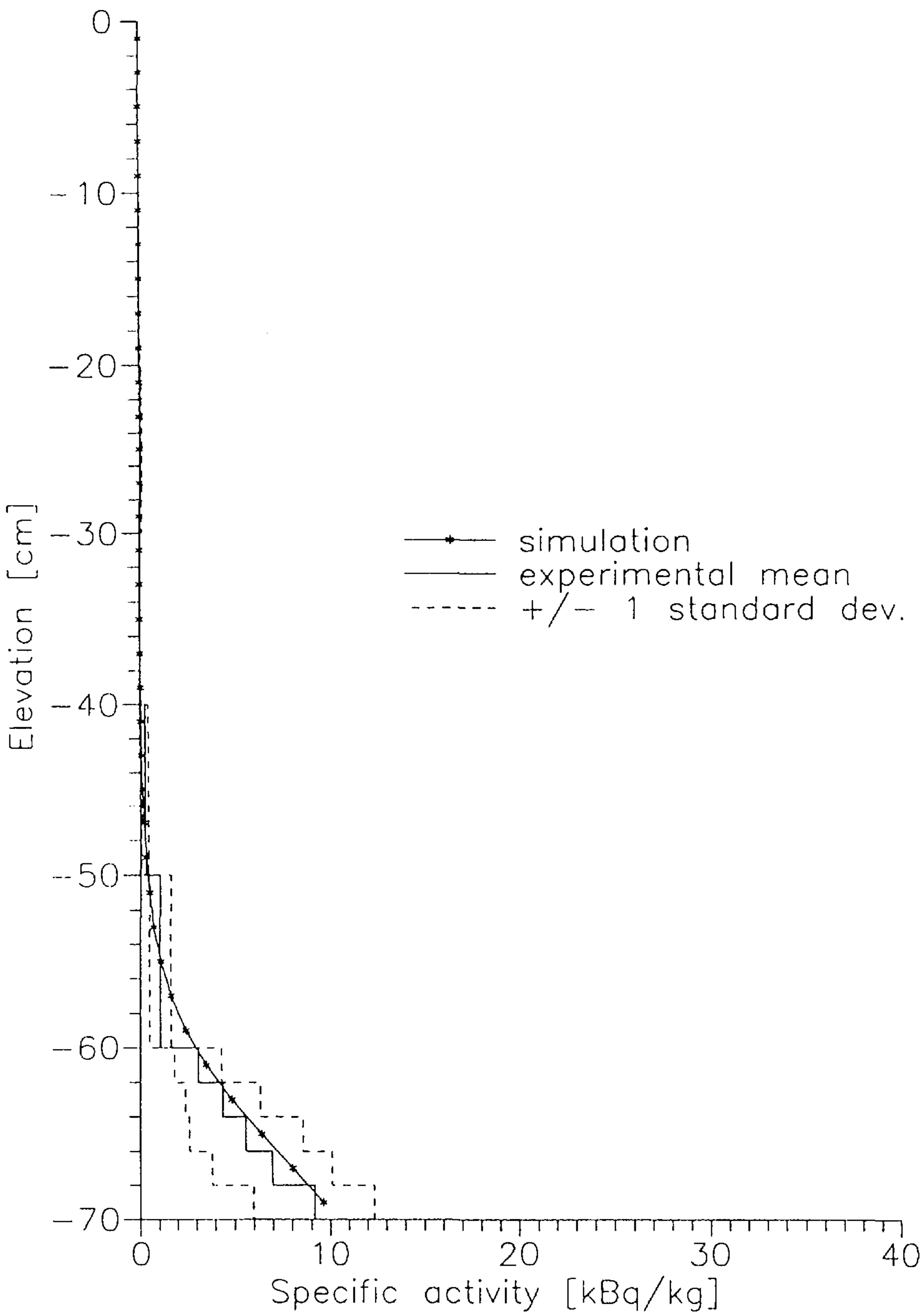

Figure 5a: Shallow lysimeter specific activity depth profile Julian Date 92209

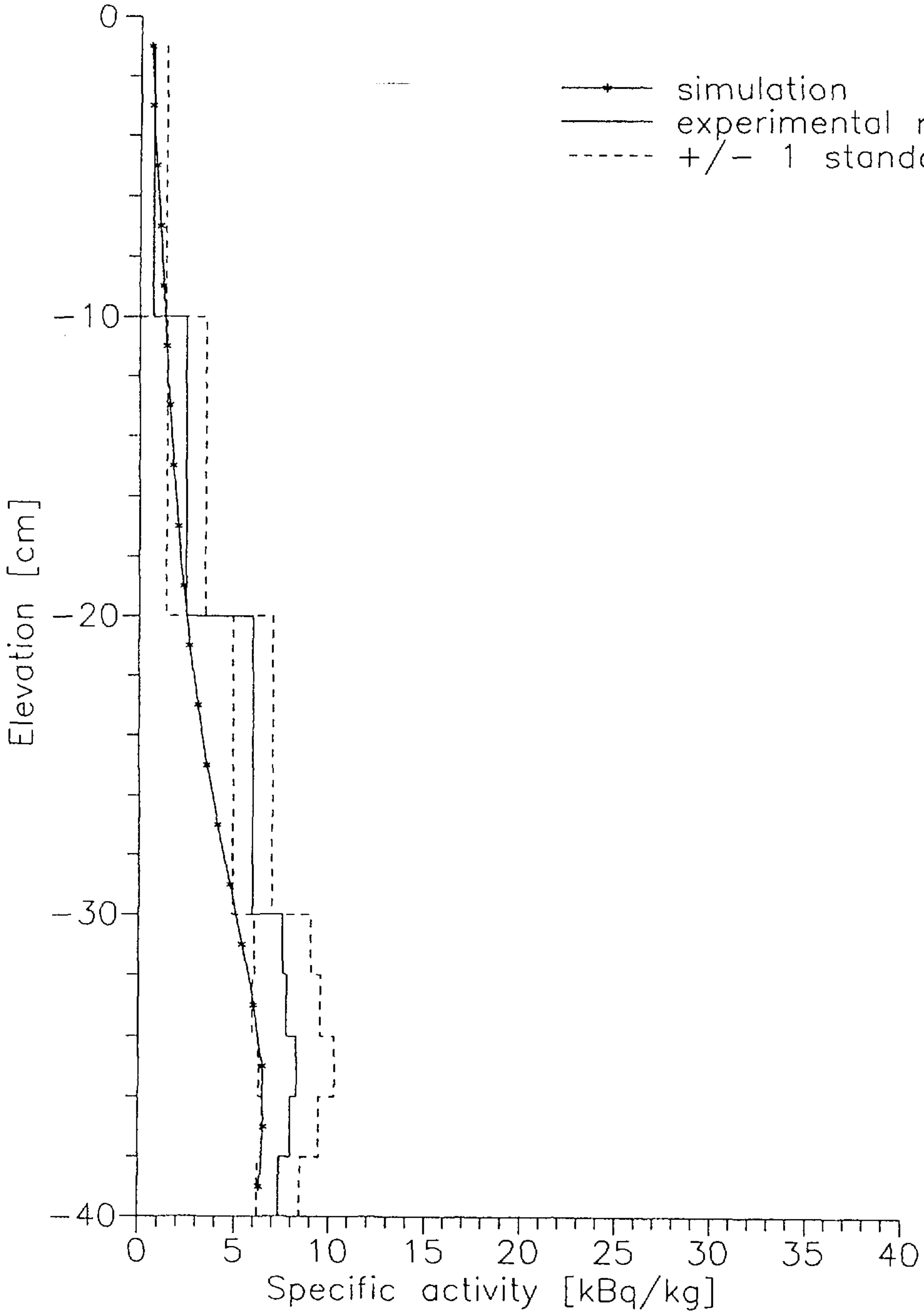

Figure 5b: Deep lysimeter specific activity depth profile Julian Date 92209

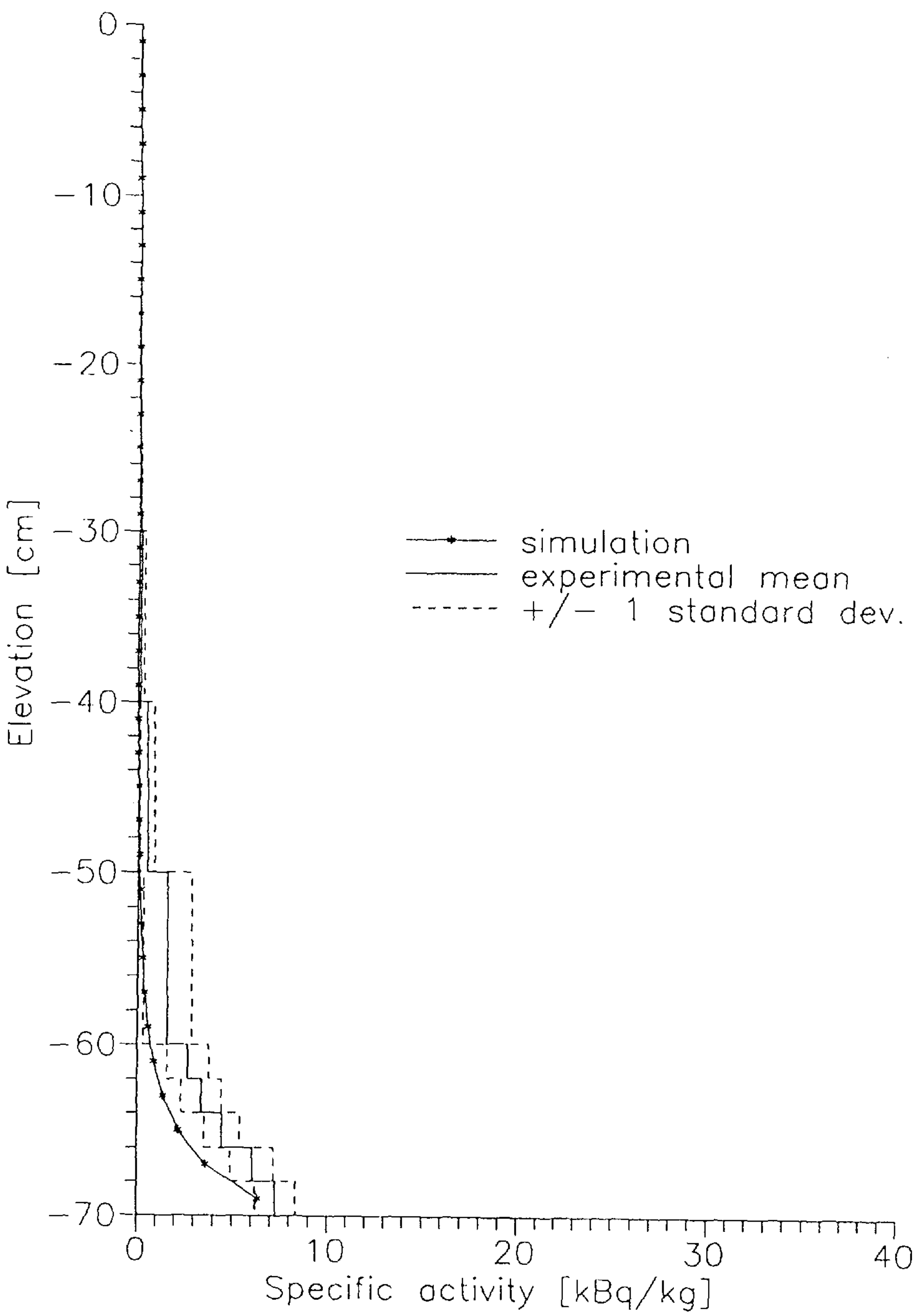

Figure 6: ^{22}Na TF_w values in grain, chaff, leaf and stem in both deep and shallow lysimeters in 1990, 1991 and 1992.