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# Assessment of spatial redistribution of Chernobyl-derived radiocaesium within catchments using GIS-embedded models

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**Abstract** The Chernobyl accident has resulted in surface contamination by radiocaesium ($^{137}$Cs) over vast areas of eastern and northern Europe. This surface contamination has been subject to changes due to physical decay and lateral transport of contaminated soil particles, which has resulted in a still on-going transfer of radionuclides from terrestrial ecosystems to surface water, river bed sediments and flood plains. Evidence from previous research show that this may cause a local enhancement of $^{137}$Cs uptake into food chains. Although $^{137}$Cs has been used as tracer in many soil erosion modelling studies, spatially distributed erosion and sedimentation models have not been widely used for evaluation of radionuclide transport within and from river catchments. This paper presents an integrated set of GIS-embedded models that are being developed in the framework of the EC-financed project SPARTACUS to assess redistribution of radionuclides at the catchment scale. These models are implemented in the spatio-temporal modelling language of the raster-GIS PCRaster and account for water runoff, soil erosion and deposition, and sediment-associated radionuclide transport.

## INTRODUCTION

After the accident at the Chernobyl nuclear power plant on 26 April 1986, vast areas of eastern, central and northern Europe were contaminated by $^{137}Cs$ (Fig. 1). The initial $^{137}Cs$ deposition patterns were determined by dispersion and deposition processes governed by meteorological conditions during the first days after the accident. The territories of Ukraine, Belarus, and the European part of Russia were most affected. Nevertheless, areas farther away from Chernobyl were also affected by relatively high levels of deposition due, but not solely, to rainfall as the released radioactivity passed over these areas (De Cort *et al.*, 1998).

In the years following the Chernobyl accident, surface contamination by $^{137}Cs$ has been subject to changes not only due to physical decay, but also from lateral transport of contaminated water and soil particles. The mobility and fate of $^{137}Cs$ in landscapes is largely determined by its geochemistry. Caesium-137 is very soluble in water, but also readily adsorbed by illitic clay minerals (Cremers *et al.*, 1988). As a consequence, in mineral soils $^{137}Cs$ becomes irreversibly fixed to clay minerals over time, whereas in organic soils it remains mobile, which has been demonstrated by many authors (Hilton *et al.*, 1993; Absalom *et al.*, 1995; Kudelsky *et al.*, 1996).

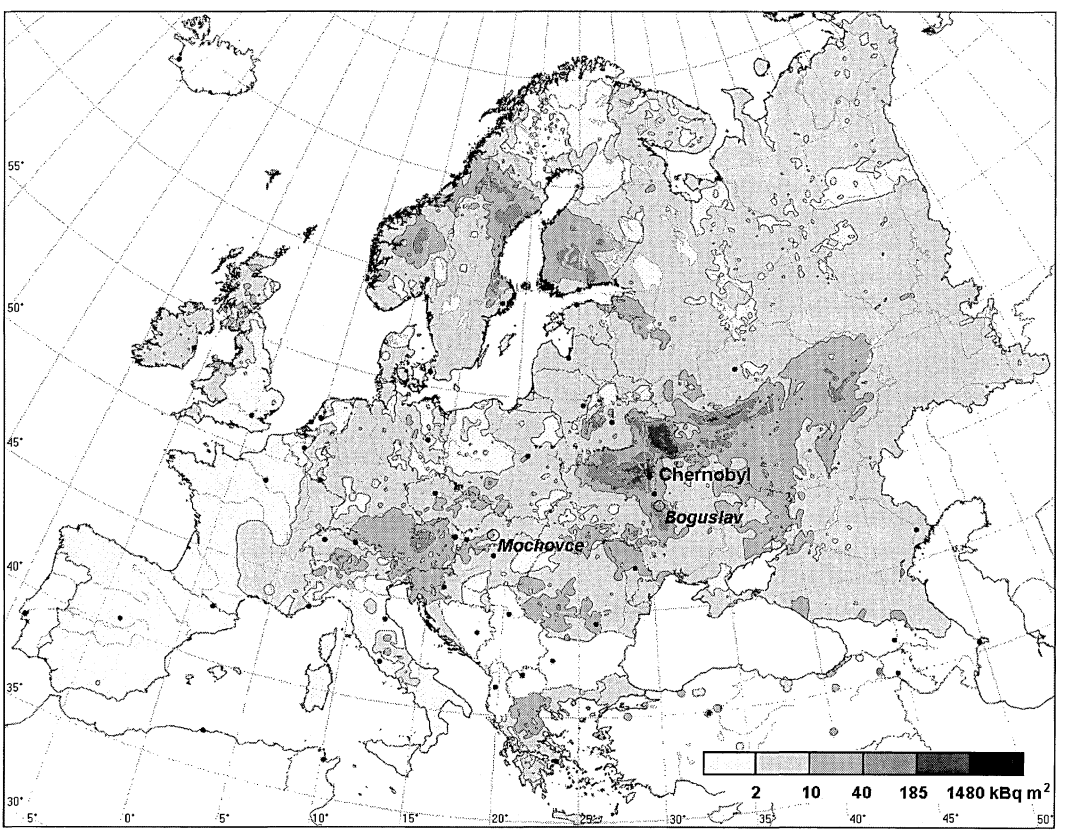


**Fig. 1** Caesium-137 deposition in Europe (May 1986) (kBq m$^2$) (source: De Cort *et al.*, 1998). The study catchments (Mochovce, Slovakia and Boguslav, Ukraine) are indicated by O.

Hence, the transport of $^{137}Cs$ from and redistribution within agricultural and natural catchments in the long term is mainly related to the transport of fine sediments, although the initial post-accidental transport of $^{137}Cs$ might largely have occurred in dissolved form (Hilton *et al.*, 1993; Slavik *et al.*, 1997).

Secondary contamination of surface water due to exchange between highly contaminated flood-plain soils and river water is an important issue in the most contaminated areas around Chernobyl (Sansone & Voitsekhovitch, 1996). In these areas, additional inputs of $^{137}Cs$ via deposition of contaminated sediment from upstream catchment areas is considered to be negligible compared to the initial $^{137}Cs$ deposition inventory, whereas in areas known as "relatively minor contaminated areas", local enhancement of $^{137}Cs$ inventory due to deposition of contaminated sediments on flood plains may be relevant. Moreover, hydrochemical changes in submerged flood-plain soils during flooding may influence transfer rates from soil to vegetation. Recent studies have shown that this may cause a local increase of $^{137}Cs$ uptake into food chains (Van der Perk *et al.*, 1999b). Therefore, it is essential to consider redistribution processes that affect the initial $^{137}Cs$ deposition patterns for an appropriate assessment of pathways by which radioactivity is transferred in ecosystems.

## SCOPE OF THE PROJECT

Most modelling studies on radionuclide redistribution have focused on the transfer from terrestrial to aquatic ecosystems (Shukla, 1993; Monte, 1995; Sansone & Voitsekhovitch, 1996; Vakulovski *et al.*, 1996). These studies have considered only dissolved radionuclide transport and have yielded aggregated predictions at the scale of entire catchments. Spatially distributed erosion and sedimentation models have not been widely used for evaluation of transport within and from river catchments (Popov, 1995), although $^{137}Cs$ has been used as tracer in many soil erosion modelling studies (de Roo & Walling, 1994; Walling *et al.*, 1995; Walling & He, 1997).

The main aim of the EC funded SPARTACUS project (EC DGXII INCO-COPERNICUS Programme) is to develop an integrated set of distributed models for assessment of the spatial redistribution of $^{137}Cs$ within catchments. These models account for both solute and sediment associated transport as a result of surface runoff, soil erosion and deposition at various time scales. The redistribution models improve our estimations of changes in local soil contamination by $^{137}Cs$ compared to merely a correction of initial deposition for physical decay. Simultaneously, spatial patterns of surface water contamination by $^{137}Cs$ are predicted.

## GIS-EMBEDDED MODELS OF CAESIUM-137 REDISTRIBUTION

### GIS-embedded modelling

Recent developments in Geographical Information Systems (GIS) and digital elevation models have enabled the construction of hydrological, soil erosion, and water quality models embedded within a GIS environment (Burrough, 1996). The

advantage of embedded coupling of models to GIS is the accomplishment of a full integration of the spatial database and the model, which facilitates the development and adaptation of models and exploration of model results. The spatio-temporal modelling language of the raster-GIS PCRaster (Wesseling *et al.*, 1996) offers a convenient framework to build the GIS-embedded $^{137}Cs$ redistribution models, providing a powerful decision support tool. This approach has been successfully applied in the development of an environmental decision support system (EDSS) for the assessment of $^{137}Cs$ transfer through food chains in areas contaminated by the Chernobyl accident (Van der Perk *et al.*, 1998, 1999a). The models being developed within the framework of the SPARTACUS projects will be integrated within this EDSS and, therefore, implemented into the PCRaster GIS.

Prediction of the changes in soil and surface water contamination by $^{137}Cs$ is made at two time scales, namely at the event scale (hours) and the long-term scale (years). Both models are based on existing soil erosion models coupled to a $^{137}Cs$ exchange model that simulates the interaction between dissolved and adsorbed $^{137}Cs$. The basic input for both models is a map of initial soil contamination by $^{137}Cs$, a digital elevation model (DEM) and maps of soil type and land use.

## Outline of the event-based model

The event-based $^{137}Cs$ redistribution model has been based on the existing Limburg Soil Erosion Model (LISEM), a deterministic, dynamic, spatially distributed, hydrological and soil erosion model (De Roo *et al.*, 1996). Figure 2 shows the structure of the model. The LISEM model accounts for rainfall, interception, surface storage, infiltration, overland flow, channel flow, detachment by rainfall and throughfall, detachment by overland flow, and transport capacity of the soil. In addition to these processes, $^{137}Cs$ exchange processes between the dissolved and adsorbed phases in both topsoil and suspended matter have been incorporated based on a distribution coefficient approach. The distribution coefficient *Kd* ($m^3\ kg^{-1}$) is defined as the ratio between the $^{137}Cs$ activity concentration adsorbed to soil particles and/or suspended matter (Bq $kg^{-1}$) and the $^{137}Cs$ activity concentration in solution (Bq $m^{-3}$). The value for *Kd* depends on sediment type (inorganic and organic cation exchange capacity) and time due to irreversible fixation to clay minerals (Absalom *et al.*, 1995). At the event time scale, it can be assumed that *Kd* is constant for the different sediment types and equilibrium is reached instantaneously (Popov, 1995). Caesium-137 exchange processes between the topsoil and runoff water are modelled assuming an active layer of 1 mm (Popov, 1995). The initial $^{137}Cs$ contamination values usually expressed as Bq $m^{-2}$ are converted to activity concentrations in this active layer (Bq $kg^{-1}$) using a standardized depth distribution of $^{137}Cs$ and soil bulk density values based on field measurements or literature values.

## Outline of the long-term model

The long-term model consists of two sub-models for particulate $^{137}Cs$ transport on the one hand and dissolved $^{137}Cs$ transport on the other. The particulate $^{137}Cs$ transport

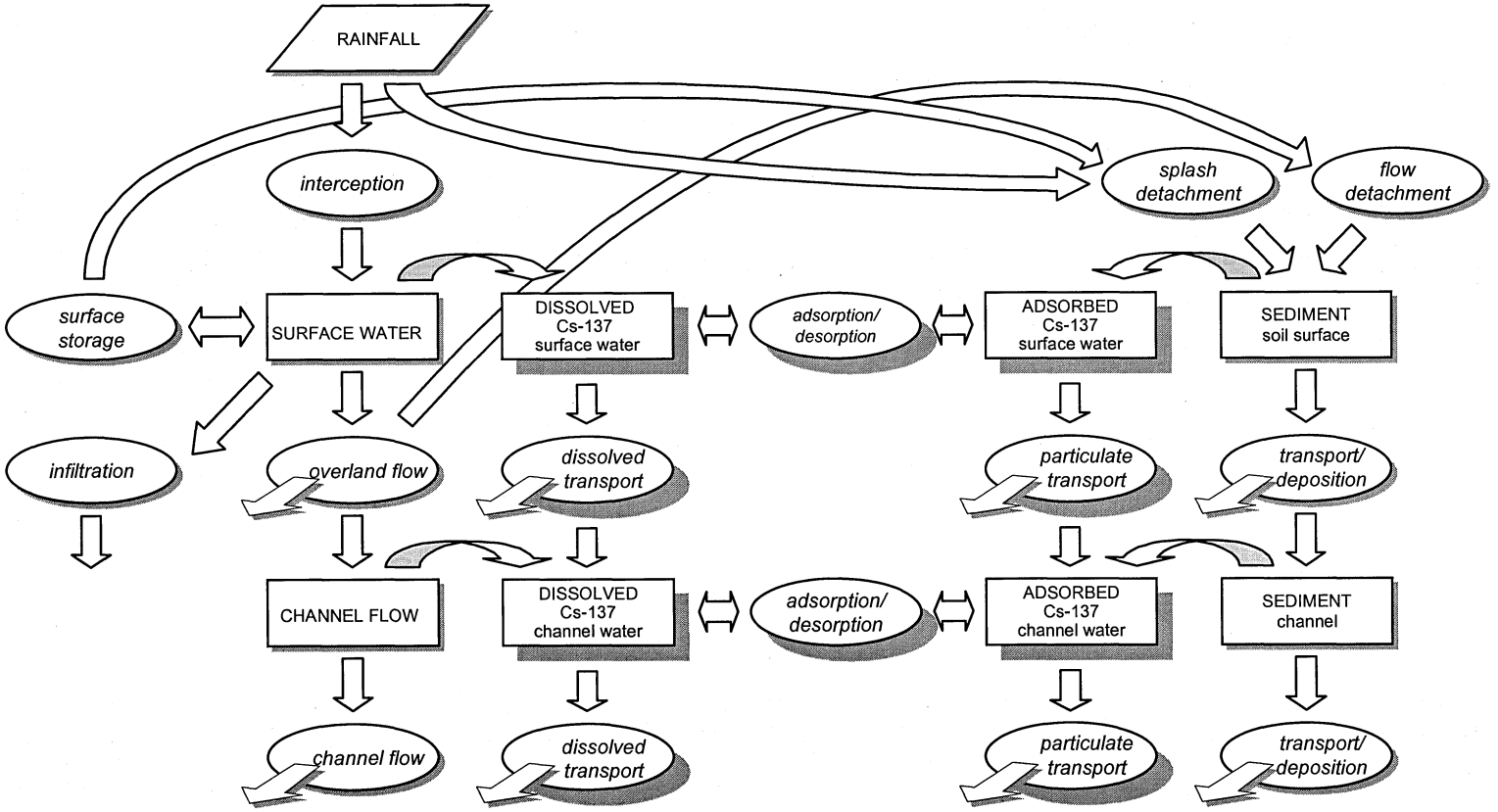


**Fig. 2** Schematic overview of the event-based radiocaesium transport model.

sub-model has been based on the rill erosion and sediment transport model described by Govers *et al.* (1995). A sediment budget index is calculated using slope gradient and slope length derived from the DEM. The logtransformed sediment budget index accounting for erosion and deposition by runoff water, as well as the slope convexity accounting for soil redistribution by tillage are entered in a linear regression model to predict actual $^{137}$Cs inventory values obtained for bulk soil cores. These predictions enable to calculate annual $^{137}$Cs redistribution rates that can be interpolated using the DEM derivatives as external variables. The resulting map of $^{137}$Cs redistribution rates is employed to predict future changes in soil contamination by $^{137}$Cs.

The submodel for long-term dissolved $^{137}$Cs transport is based on a lumped radionuclide wash-off model described by Smith *et al.* (in press). In this approach, $^{137}$Cs transport described by a wash-off coefficient, which is defined as the ratio between the $^{137}$Cs activity concentration in runoff water and the total $^{137}$Cs inventory in the catchment. The time dependence of the wash-off coefficient is modelled using a series of exponential functions (Monte, 1995):

$$R = Ae^{\lambda_1 t} + Be^{\lambda_2 t} + Ce^{\lambda_3 t} \quad (1)$$

where $R$ is the radiocaesium wash-off coefficient (m$^3$ kg$^{-1}$); $A$, $B$, $C$ are empirical constants (-); and $\lambda_1$, $\lambda_2$, $\lambda_3$ are $^{137}$Cs decline constants representing the fast, intermediate and slow pool (year$^{-1}$).

The wash-off coefficient depends on catchment soil type, which probably explains the variation in wash-off coefficients for different catchments (Smith *et al.*, in press). Catchments with large coverages of peat soils are likely to have significantly higher wash-off coefficient values. The implementation of this lumped approach into a distributed GIS model has overcome this problem by assigning calibrated wash-off coefficient by soil texture classes. The $^{137}$Cs activity concentration in runoff water is calculated from the product of the wash-off coefficient and $^{137}$Cs inventory derived from the particulate $^{137}$Cs transport model. Subsequently, transport is calculated by multiplying the activity concentrations with spatially distributes estimations of water runoff.

## DATA COLLECTION

A field data collection programme has been initiated to provide data for the above described GIS-based models. For this purpose, several study catchments have been selected, which have been monitored for $^{137}$Cs contamination since the Chernobyl accident. These catchments include the Boguslav experimental catchments within the Butenya River basin (59 km$^2$) about 100 km south of Kiev, Ukraine, and the Mochovce experimental catchment (3.7 km$^2$) in western Slovakia (see Fig. 1). These catchments are being investigated during 1999 and 2000. Besides a programme to obtain basic model input maps of soil type, land use, and a detailed DEM, sampling networks have been set up to measure actual soil contamination by $^{137}$Cs. These $^{137}$Cs measurements will be related to DEM derivatives in order to improve the interpolation of point samples to continuous surfaces.

The event-based $^{137}$Cs transport model needs input of various soil properties, such as infiltration capacity, soil bulk density, porosity and texture, initial soil

moisture, surface roughness, and cohesion. To obtain maps of these parameters, field observations were made and will be made along various transects through the study catchments, which will be interpolated using the basic input maps. In addition, field estimates of interception parameters (leaf area index) were made and the number and location of channels, tracks and furrows were recorded.

Furthermore, detailed information at the event scale was and will be collected to calibrate and test the models. For this purpose, during rainstorm events the study catchments are being monitored with respect to rainfall, runoff and $^{137}Cs$ activity concentration in both the dissolved and adsorbed phase at the catchment outlets.

## CONCLUDING REMARKS

The SPARTACUS project has provided a unique opportunity to apply the novel fields of GIS and radio-ecological modelling to existing and modern experimental data sets on radionuclide redistribution. The models outlined in this paper combine existing modelling approaches for soil erosion at the catchment scale with approaches for modelling exchange of $^{137}Cs$ dissolved in runoff water and adsorbed to soil or suspended solids. The execution of these models within the raster GIS environment accomplishes an integral coupling between the geographic databases and the various models at different time scales. Accordingly, the integration of these models within an environmental decision support system provides a power tool to assess the radio-ecological effects of $^{137}Cs$ redistribution, which can be adjusted easily to the user's needs. Through such an integration, a new basis for development of spatial decision support tools for radionuclide transport control at the catchment scale in radioactively affected areas is achieved. Since the collection and processing of field data is in progress, modelling results have not been available yet, but will be published in separate papers in due time.

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