# Downward Migration of Chernobyl-derived Radionuclides in Soils in Poland and Sweden

Gerald Matisoff[1], Lauren F. Vitko[1], Peter J. Whiting[1], Michael E. Ketterer[2], Jerzy W. Mietelski[3], Edyta Lokas[3], Klas Rosen[4], and Henning Persson[4]

*[1]Case Western Reserve University, Cleveland OH 44106-7216 USA, [2]Northern Arizona University, Flagstaff AZ 86011-5698 USA, [3]The Henryk Niewodniczanski Institute of Nuclear Physics, Krakow Poland, [4]Swedish University of Agricultural Sciences Uppsala Sweden*

## INTRODUCTION

The Chernobyl accident on April 26, 1986 resulted in significant fallout of radionuclides such as $^{137}$Cs and $^{239,240}$Pu on surface soils throughout northern Europe. Knowledge of the dynamics and mechanisms of the migration of these radionuclides in soils is important for determining animal and human dose exposure rates, determining exposure from food-chain transfer, and in planning environmental remediation and clean-up. In addition, both natural ($^{7}$Be, $^{210}$Pb$_{xs}$) and anthropogenic ($^{137}$Cs, $^{239,240}$Pu) fallout radionuclides have been extensively employed to determine short-term soil erosion rates, to trace sediment source regions, to characterize and quantify erosion mechanisms, to constrain sediment budgets, and to better understand the delivery ratios, transit distance and transit time of fine sediment and adsorbed pollutants. Despite the use of these radionuclides and their activity-depth profiles, little is known about how the profile shapes develop or why they differ with location. There is a substantial amount of information that is embedded in the distribution of radionuclides with depth that could be extracted to refine our ability to understand significant radiochemical behavior, to predict dose exposure rates, to better plan environmental remediation, to use these and other radionuclides to understand soil erosion mechanisms, to identify sediment source areas and to calculate watershed inventories and residence times for better understanding of watershed retention and erosion processes. Moreover, ignoring this evidence may lead to a real risk of drawing incorrect conclusions from an incomplete understanding of the radionuclide profiles.

The primary goal of the research is to characterize how radionuclides migrate through soils and attach to fine sediment and are eroded from the landscape and transported through stream networks. At the same time the findings have implications for questions of human and ecosystem health, landscape evolution, geochemical cycling and water resource degradation. The downcore distributions of natural and anthropogenic radionuclides in soils are controlled by a variety of processes, including depositional flux, precipitation, infiltration, porosity/permeability, water content, dispersion coefficient, organic matter content, clay content, bulk density, particle size, adsorption, cation exchange capacity, colloidal transport, bypass flows through cracks, bioturbation and radioactive decay. The objective of this research is to develop a better quantitative understanding of the processes that lead to the development of radionuclide profiles in soil columns. To help constrain the results, the work was conducted at a series of field locations that cover a range in values for several key parameters. Presented here are soil profiles of $^{137}$Cs, $^{210}$Pb$_{xs}$ and $^{239,240}$Pu from the Lazy, Tulowice and Bor za Lasem sites in Poland and the Hille and Skogsvallen sites in Sweden so that the profiles of these nuclides may be compared with those obtained previously (Chemicki et al., 1996-1997; Boro et al., 2001; Mietelski et al., 2004; Rosen et al., 1999) to better understand the downward migration.

## MATERIALS AND METHODS

### Study area

The Lazy, Tulowice and Bor za Lasem sites in Poland and the Hille and Skogsvallen sites in Sweden were selected for sampling because of previous work at the locations. The study sites exhibit a range of properties that affect radionuclide retardation or migration and at which there are prior radionuclide profiles to provide a time series of data that can be used to constrain the modeling. The study sites span a range of geography, elevation, precipitation, Chernobyl fallout flux and soil orders. In particular, Bor za Lasem is a peat and is a relatively unique situation compared to mineral soils – it might be expected to have very unusual characteristics with poor Cs binding but possibly good Pu binding and immobilization.

### Soil Sampling

Vertical sections of the soil profiles were collected using both tube corers and our high-resolution corer (Wilson et al., 2003). Samples were collected from all sites in 2007, although six vertical profiles from each of the two sites in Sweden were collected previously over the 1987-2006 time period. We took samples from undisturbed, relatively flat sites that appeared to be neither the site of erosion nor deposition. In the upper part of the soil, the sampling interval was 0.4-3.5 cm while deeper in the cores the sampling interval ranged from 0.8-5 cm depending upon depth and type of corer. Most soil cores were collected to a depth of 25-30 cm although a few cores were collected to a depth of 60 cm. Soil cores were returned to the laboratory for processing and analysis. Soil samples were dried at 80-105º C, ground, placed in containers and sealed in preparation for radionuclide analysis.

### Gamma Spectroscopy and ICPMS

Radionuclide activities were determined by gamma spectroscopy. Gamma spectroscopy for $^{210}$Pb (46.52 keV), $^{7}$Be (477.6 keV), $^{137}$Cs (661.65 keV), and $^{214}$Bi (609.3 keV) is reasonably well-established and straightforward, but the analytical details are detector and sample specific. Among the three labs in the US, Poland, and Sweden, we used eight HPGe gamma detectors for the radionuclide analyses. Activities measured between the different detectors varied by less than 10%. Samples were counted for 2-24 hours to decrease the counting errors (~10%) associated with small samples and low radionuclide activities. For the $^{210}$Pb analyses, the soil samples were counted after three weeks to allow secular equilibrium ingrowth of gaseous $^{222}$Rn ($t_{1/2}$ = 3.82 d) from the decay of its $^{226}$Ra ($t_{1/2}$ = 1600 yr) parent. The self-attenuation correction of Cutshall et al. (1983) was used. Standards were prepared using the same geometries as the samples. The instrumentation and counting geometries were calibrated using commercially available mixed energies gamma-emitting standards. All measured counts were corrected for background levels, detector and geometry efficiencies, branching ratios and decay.

Plutonium activities ($^{239+240}$Pu) and atom ratios were determined in $HNO_3$/HF digested soils following the methods described in Cizdziel et al. (2008). $^{242}$Pu (0.007 Bq) was used as a spike isotope; Pu fractions were processed using TEVA resin, and Pu isotopes were measured by sector field ICPMS.

### Computer Models

Preliminary calculations were performed using the model CHAIN (van Genuchen, 1985) and HYDRUS-1D (Simunek, Van Genuchten, and Sejna, 2005) to illustrate the nature of solute migration in a soil profile and how model simulations can provide insight to the key processes governing transport.

## RESULTS AND CONCLUSIONS

The $^{137}$Cs fallout by Chernobyl is about 30 times greater than that from bomb fallout at the three sites in Poland and about 300 times greater than at the two sites in Sweden. Vertical profiles of

$^{7}$Be, $^{210}$Pb, and $^{137}$Cs activities in soils indicate that $^{7}$Be activity decreases rapidly downcore due to its short (53 d) half-life. Excess $^{210}$Pb from atmospheric fallout decreases downcore more slowly due to its longer (20.4 yr) half-life. Some $^{210}$Pb activity persists at depth because of *in situ* production from $^{226}$Ra decay. $^{137}$Cs activity displays a subsurface peak reflecting Chernobyl fallout and subsequent downcore migration. A time series of the $^{137}$Cs profiles indicates that the peak in $^{137}$Cs activity occurred at the soil surface in 1987, but migrated downcore about 4 cm since, and that some $^{137}$Cs activity can now be found as deep as 20 cm. The amount of downward migration is different at the different locations. This is likely due to the different types of soils, where some soils are dominated by mineral matter and others by organic matter. At Hille at a depth of 32 cm there is a second, small peak that might be the 1963 fallout peak. Work currently in progress is using Pu atom ratios to distinguish between Chernobyl and stratospheric fallout. The $^{240}$Pu/$^{239}$Pu atom ratio is about 0.38 from Chernobyl and 0.18 from stratospheric fallout. There are also even larger relative differences in $^{241}$Pu/$^{239}$Pu and $^{242}$Pu/$^{239}$Pu which can be measured if the activities are high enough. It is also expected that differences exist in micromineralogy and phase association between stratospheric fallout and Chernobyl Pu; the latter is primarily associated with refractory U-rich fuel particles.

CHAIN model simulations capture some of the major features of the $^{7}$Be, $^{137}$Cs, and $^{210}$Pb$_{xs}$ profiles. The simulations generate profiles that decrease downcore for both $^{7}$Be and $^{210}$Pb$_{xs}$ while also accounting for the differences in half-life of these two radionuclides. The $^{7}$Be migrates only about 1 cm before its activity approaches 0 whereas $^{210}$Pb decreases throughout the 25 cm that were simulated. The continuous input of $^{210}$Pb$_{xs}$ results in a profile that remains fixed in time whereas a pulsed input for$^{137}$Cs results in a downward migration of the $^{137}$Cs peak with time. Dispersion results in tails on both the leading edge and the trailing side of the $^{137}$Cs pulse and the importance of sorption and retardation is easily seen in both the $^{137}$Cs and $^{210}$Pb profiles. However, the CHAIN model fails to effectively describe three aspects of the data: 1) the short term transport as seen in the $^{7}$Be data is deeper than the model simulations would suggest; 2) some of the $^{137}$Cs and $^{239,240}$Pu have been retained in the top portion of the core whereas the model predicts that these radionuclides will be transported downcore as a pulse; and 3) $^{210}$Pb activities decrease more rapidly near the surface than the model would predict.

CHAIN and other advection-dispersion models of solute transport fail to accurately describe some aspects of the downward migration of radionuclides in the unsaturated soil zone. HYDRUS-1D has the potential to better describe the processes by incorporation of nonlinear adsorption, inclusion of soil properties into multiple zones, and accounting for time-dependent processes such as water content. Future work will involve additional modeling using the program HYDRUS-1D which should permit exploring the causes for these differences to refine our ability to describe the transport of radionuclides in soils, for example, by permitting the use of nonlinear adsorption or by defining soil properties into multiple depth zones.

## ACKNOWLEDGEMENT

This work was supported by the Office of International Programs at NSF, grant number OISE-0745066.

## REFERENCES


K. Boron, J. W. Mietelski, K. Lipka, P. Gaca, M. Jasinska, 2001. Radionuclides in raised bogs on example of „Bór za Lasem" in Orawsko-Nowotarska Valley in Tatra Mountains Foothill, Poland. Journal of Environmental Monitoring, 3(3:) 324-329.

Chelmicki, W., M.Klimek, K.Krzemien, M.Jasinska, K.Kozak, J.W.Mietelski, 1996-1997. Spatial distribution of Cs-137 on Pogórze Wielickie near Bochnia. Folia Geographica, Ser. Geogr. Phys., XXVIII: 35-47 (in Polish).

Cizdziel, J.V., M.E. Ketterer, D. Farmer, S.H. Faller, and V.F. Hodge, 2008. $^{239}$Pu, $^{240}$Pu, $^{241}$Pu fingerprinting of plutonium in western US soils using ICPMS: solution and laser ablation measurements. Analytical and Bioanalytical Chemistry, 390:521-530.

Cutshall, N. H., I.L. Larsen and C. R. Olsen, 1983. Direct analysis of $^{210}$Pb in sediment samples: self-adsorption correction. Nuclear Instruments and Methods, 206:309-312.

Mietelski, J.W., P.Szwalko, E.Tomankiewicz, P.Gaca, S. Malek, J.Barszcz, S.Grabowska, 2004. $^{137}$Cs, $^{40}$K, $^{90}$Sr, $^{238,}$ $^{239+240}$Pu, $^{241}$Am and $^{243+244}$Cm in forest litter and their transfer to some species of insects and plants in boreal forests - three cases study. Journal of Radioanalytical and Nuclear Chemistry, 62(3): 645-660.

Rosén, K., I. Öborn and H. Lönsjö, 1999. Migration of radiocesium in Swedish soil profiles after the Chernobyl accident, 1987-1995. J. Environ. Radioactivity, 46:45-66.

Simunek, J., M. Th. Van Genuchten, and M. Sejna, 2005. The Hydrus-1D software package for simulating the one-dimensional movement of water, heat, and multiple solutes in variable saturated media (version 3.0).

Van Genuchten, M. Th., 1985. Convective-dispersive transport of solutes involved in sequential first-order decay reactions. Computers & Geosciences, 11: 129-147.

Wilson, C.G., G. Matisoff, and P.J. Whiting, 2003. Short-term erosion rates from a $^{7}$Be inventory balance. Earth Surface Processes and Landforms, 28(9): 967-977.