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Radiocesium and ^{210}Pb in sediments, soils and surface waters of a high alpine catchment: A mass balance approach relevant to radionuclide migration and storage

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ABSTRACT

The behaviour of particle-reactive metals in natural environments is linked to adsorption, particle erosion, transport and sedimentation. In the field these processes are best studied and quantified on small, local scales, where the number of variables remains manageable. The Muttsee watershed, situated above 2500 m in the north-eastern Swiss Alps, represents such a 'simple' natural test site, a self-contained bedrock-soil-water-sediment system that allows the analysis of radioactive metals in order to study their interaction with water, bedrock, soil and sediment and to quantify radionuclide and particle transfer. A small lake (Muttsee) forms a natural trap for eroded material and ~ 80% of the radionuclides ^{210}Pb and ^{137,134}Cs deposited in the catchment area. Radionuclide-dated sediment cores from the lake allow the determination of sedimentation rates, which increased from 0.30 cm year^{-1} during the period 1963–1986 to 0.55 cm year^{-1} during 1986–1992. The related increase in erosion rate (10 to 19 cm kyrs^{-1}) may be related to a landslide that occurred during the early 1960s, when sections of plant-stabilized soil were covered by rock debris. The data, which can be evaluated using mass balance calculations, allow assessment of the role of a soil cover in controlling radionuclide residence times in the drainage basin, and also allow estimation of changes in the extent of soil cover during the last 40 years. This changing extent of soil cover makes it impossible to use soil profiles for an assessment of atmospheric radionuclide fluxes. Radionuclide measurements in soils serve as a pedological tool, allowing in cases recognition of soil creep and transient cover.

Introduction

A clear understanding of radionuclide migration in geologic media is complicated by a myriad of physical and chemical processes that play important roles in natural environments. The pressing need to further investigate this complex matter becomes obvious by screening publications dealing with nuclear accidents and spills (Aarkrog, 1988; UNSCEAR, 1993) and by the necessity to evaluate pre-storage radionuclide levels of nuclear waste disposal sites. In Central Europe such conta-

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mination is mostly due to radionuclide fall-out from atmospheric atomic bomb testing and the accident in Chernobyl (Devell et al., 1986; EML, 1977; Völkle et al. 1989).

I have chosen an inventory approach to study radionuclide migration in all relevant compartments of a watershed. Various natural processes are relevant: precipitation, run-off, weathering, erosion, transport and deposition. Most of these processes are linked to the extent and thickness of the soil and regolith cover and its stabilization by plant growth. To avoid getting lost in a labyrinth of complexities, I decided to work in a relatively simple alpine area, located in the Glarus canton of east-central Switzerland (Fig. 1). The area comprises a small lake (Muttsee, 0.42 km^2 surface area at 2500 m altitude), which forms a natural trap for all eroded particles from the 2.95 km^2 large catchment (excluding the lake).

Erosion processes have been described and quantified using denudation rate protocols, such as dating colluvial deposits (Reneau et al., 1989) or in-situ produced cosmogenic nuclides (Albrecht et al., 1993; Cerling and Craig, 1994). The application of natural (^{210}Pb) and anthropogenic (^{137}Cs) radionuclides has been intensively applied in Australia and the United States (Lance et al., 1986; McIntyre et al., 1987; Ritchie and McHenry, 1975; Wallbrink and Murray; 1993), mostly in cultivated areas and will be applied here to a watershed dominated by physical erosion.

With the goal in mind to assess the total ^{210}Pb and $^{137,134}Cs$ activity of a specific area, we incorporated the classical geomorphic technique (estimating the mass of deposited sediment and its relationship to the surface of the catchment area) with a budget for the specific radionuclides, accumulated in eroded particles (lake sediments) and in soils. The sediments in the lake can be dated based on ^{137}Cs and serve as archives for the radionuclide flux into the lake during the last 40 years. Budgeting two independent radionuclides in all input and output compartments allows a detailed evaluation of the status quo, i.e. assessing the proportion of radionuclides stored in soils vs. sediments or the flux terms between them. Possible changes in the catchment during the last half century can be extracted from the sediment archives.

Field information

Geology

A simplified geologic map (Fig. 1) outlines the major topographic and geologic units of the catchment area. The highest ridges and crests (reaching almost 3000 m a.s.l.) are formed by sandstones and wildflysch, overlying schists that form most of the steep slopes. The lower portion of these slopes surrounding the lake are covered with active talus debris. South of the lake a small plateau is formed by marl, marly limestone and sandstone. The schists are basically impermeable. A slight increase in permeability can be observed in the sandstones and even more so in the sandy marl. The site has a weathering-limited denudation regime (Stallard and Edmond, 1983). Potential mechanical erosion is considerably faster than chemical weathering due to the high altitude of 2500–3000 m and temperatures which lie below zero during three quarters of the year (c.f. Drever and Zobrist, 1992).

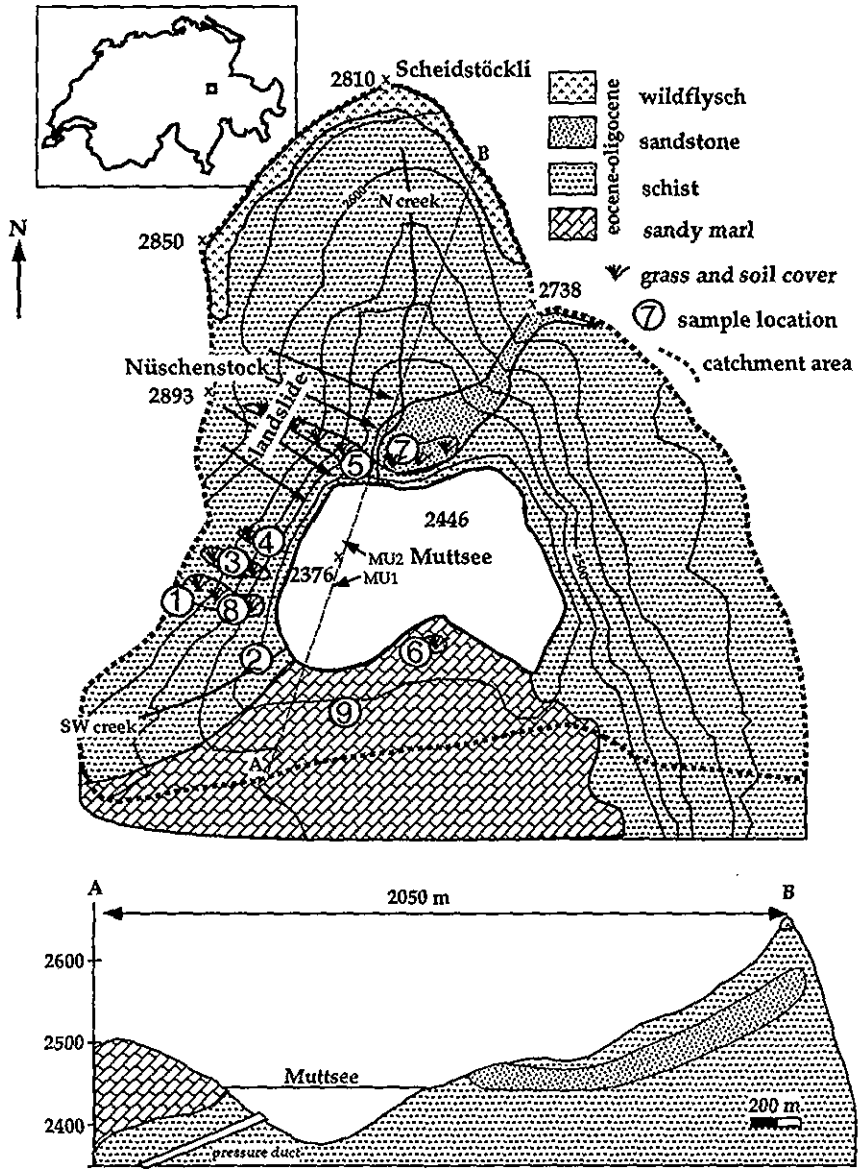

Figure 1. Generalized geologic map (above) and cross-section (between markers A and B, below), showing the major geologic units, the topography, the main creeks, the extent of soil and grass cover, the catchment area, the site of a major landslide in the early 60s and all sampling locations (MU1 and MU2 indicate the two sediment cores; rock samples at 1 relates to an unaltered schist, 3 to an altered schist, 6 to a sandy marl, 7 to unaltered sandstone; soil sections at 5, 6, 7 were taken on flat areas; 3, 4, 8 on steep areas; surface water samples were collected near 2, 5, 6, 9). The geology at depth in the cross-section is for illustrative purposes only. The inset in the upper left corner shows a map of Switzerland with the locality of the study area

Hydrology and climate

Estimated precipitation averages 1800 mm yr^{-1}, most of which falls in form of snow, at times even in the summer. The drainage basin has a surface area of 2.95 km^2, seven times larger than the surface of the lake (Fig. 1). The area is mostly free of snow in the months of July, August and September. Due to the abundance of impermeable schists in the catchment area, and the lack of a conduct system between the lake and the caves just south of the Muttsee catchment area (Weidmann et al., 1994) subterranean water loss is likely to be insignificant. Since the 1950's, natural overflow of the lake has ceased and water leaves the lake via a pressure duct used for hydroelectric purposes. Water is drained from the lake only during fall and winter months, which allows sufficient time for settling of particles washed into the lake during the warmer period. The average water volume drained through the duct system (5.5×10^6 m^3) differs from the volume entering the catchment area (6.1×10^6 m^3 yr^{-1}, for 1992) by 10%, a typical value for evaporation in high alpine regions (Lang, 1978). This supports the hypothesis of insignificant subterranean water loss. In the southern and western section of the catchment area, overland flows are common, but are quickly channelled in small ravines of which two form larger creeks that carry water on a more regular basis. In the northern and eastern portion, precipitation flows through the vadose zone of alluvial fans to reach the lake. A single surface water measurement yielded a pH of 6.0.

Pedology

Due to the blocky character of the talus debris, the east and west side of the lake catchment area are barren of regolith and soil. Slaty mud accumulates on the west side of the catchment area. Some low ridges separating ephemeral channels sustain thin soil covers. The existence of a soil cover on steep walls at this altitude is the result of deposition of sediment scavenged off a larger rock surface. Most of the presently active surface flow interacts in a complex fashion with these steep soil covered areas. Smaller erosion protected locations north and south of the lake support an additional patchy soil cover (Fig. 1) and interact with surface run-off originating in a large area. The total section covered by soil is approximately 82 000 m^2, which represents 2.8% of the catchment area (excluding the lake). The soil is young and lacks horizonation, its thickness varies between 10 and 30 cm and densities between 0.4 and 0.7 g/cm^3. The plant cover is sparse, dominated by different grasses, moss and shrubs not taller than a few cm.

Radionuclide deposition

Atmospheric fluxes of ^{210}Pb (in Bq m^{-2} yr^{-1}) have been determined in lakes (160 for Lake Zürich, Schuler et al., 1991; 180 for Lake Sempach, Wieland et al., 1993), in ice cores (20 in Colle Gnifetti, Gäggeler et al., 1983) and directly from precipitation (140, Schuler et al., 1991). ^{210}Pb fluxes measured in ice cores tend to be too low due to snow loss during wind drift, and results obtained from lake studies are overestimates due to lateral input of ^{210}Pb. Direct flux determination for the Muttsee

area is not available. 140 Bq m^{-2} yr^{-1} is adopted here as the best estimate, even though it was measured at lower altitude. More information regarding this choice of atmospheric deposition can be found in the next section.

A total ^{137}Cs fallout inventory for atmospheric bomb testing (maximum in 1963) has been calculated by Wan et al. (1987) based on deposition of ^{90}Sr in New York (EML, 1977) and a decay corrected ^{137}Cs /^{90}Sr ratio of 1.5. Their estimate of 5.5 kBq m^{-2} is adopted here, but it should be noted that this may be inaccurate. The ^{137}Cs fall-out data (in kBq m^{-2}) as a result of the 1986 Chernobyl accident are more reliable and are given for the Dübendorf area (4.0), and for Lake Zürich (3.8; Santschi et al., 1990). A value of 2.3 kBq m^{-2} is used as the best estimate for the study area based on these values and the distribution of ^{137}Cs deposition in Switzerland (KUeR, 1989). The ^{134}Cs deposition can be obtained from the known $^{137}Cs/^{134}Cs$ ratio, 1.8 during Chernobyl fall-out in Switzerland (KUeR, 1986; Murith and Gurtner, 1995).

Radionuclide deposition values that are based on measurements carried out in the Swiss lowlands may be questionable because of higher average precipitation at elevated altitude. A detailed analysis of a soil section taken on an end moraine in the Mont Blanc Massiv (Val d'Arpette, SW Switzerland; Geering et al., 1997) clearly outlines the difficulty of choice. The site is located at 2000 m altitude, receives an average precipitation of >2000 mm yr^{-1}, and is part of the area which received according to published estimates <3 kBq m^{-2} (KUeR, 1986). The total measured soil inventory of 3.1 kBq m^{-2} (thus for Chernobyl less than 3 kBq m^{-2} due to an atomic bomb related fraction) confirms the earlier estimation, but indicates the inconsistency of a simple precipitation-deposition hypothesis. Lake Zürich which receives an average precipitation of 1100 mm/year had a higher radiocesium deposition than the Mont Blanc site with an average precipitation of >2000 mm/yr. I have therefore chosen not to make precipitation adjustments for published radionuclide deposition data, but to use the best available data set in the case of ^{210}Pb and an estimation based on the regional distribution of radiocesium. The reliability of this choice will become clear during data discussion.

Materials and methods

Radionuclides in reservoir sediments

Two sediment cores were taken on September 12, 1992 in the deeper parts of the lake (MU1, MU2, Fig. 1). Both cores were cut vertically and one side was used for sedimentologic description (Albrecht et al., 1995). The two cores were sedimentologically similar. We regard them as representative for the entire lake. Water content was determined in one core by weighing and freeze-drying. Between 3 and 8 g of dry sediment from both cores were used to measure radionuclides by γ-spectroscopy using the lines 604.7 KeV and 795.5 KeV for ^{134}Cs (2.06 years half-life), 661.7 keV for ^{137}Cs (30.0 years half-life), 46.5 KeV for ^{210}Pb (22.3 years half-life) and 477 keV for ^{7}Be (53.4 days half-life). Using a well detector shielded with an anticoincidence proportional chamber (Vojtyla et al., 1994) enables significant background reduction and thus better counting statistics. The supported ^{210}Pb activities

for core sediments were estimated on the basis of ^{210}Pb from the oldest samples in the cores. The organic content of sediments is based on organic carbon and nitrogen concentrations. Total C and N were determined using the CHNS Analyser EA 1108 built by Carlo Erba. Inorganic C is presumed to be mostly $CaCO_3$, which is obtained coulometrically. Organic C is calculated by subtracting the inorganic portion from the total. Grain-size was determined on samples of one core using the X-ray based Sedigraph 5100 (Micromeritics).

Radionuclides in soil and bedrock

Soil blocks were collected and sampled on September 2, 1993 and October 16, 1994 at 6 locations (Fig. 1) down to the C-horizon (regolith dominated by rock debris). Soil thickness varied between 10 and 35 cm . Sites #5, #6 and #7 are on flat terranes, #3, #4 and #8 on steep slopes. Plant growth on the flat areas is dominated by moss, steeper slopes are characterized by the coexistence of moss and grass (grass roots penetrating tens of cm). Soil densities were determined by excavation of soil blocks extending to bedrock, and subsequent volume and dry weight measurement. Soil samples were sectioned (mostly cm-wise) and freeze-dried. Radionuclide determination by γ-spectroscopy followed as outlined in the previous paragraph. In-situ formed ^{210}Pb was determined by γ-measurement of unaltered bedrock samples collected from each lithologic unit present in the catchment area, with the exception of the wildflysch. Even though supported ^{210}Pb in soils is usually lower than in the source rock, the errors introduced are insignificant, due to the low relative importance of the supported fraction.

Radionuclides in water samples

Water samples were collected on September 2, 1993 and October 13, 1995 in 25 and 5 liter polyethylene water containers at 5 different sampling locations (Table 2, Fig. 1). Lake water was sampled at 30 m depth via the pressure duct. Two surface run-off samples were taken from the southwestern creek, one from the northern creek and two representing surface run-off from the southern plateau (sites 6 and 9). To reduce radionuclide loss to the walls of the sample container the water samples were acidified with 40 ml 10 M HCl. In addition $FeCl_3$ and known amounts of Pb and Be salts were added to each sample as carriers. In the laboratory, after two days of equilibration, the pH was increased to 8 by adding reagent grade NH_4OH. Iron(III)-hydroxide precipitated and served as a transport phase for radionuclides in solution or adsorbed to colloids. The solutions were given two days for equilibration and hydroxide sedimentation before decantation and freeze-drying. This method is unsuitable for dissolved Cs, but applicable for colloidal Cs. Determination of the yield for ^{210}Pb and ^{7}Be extraction is based on comparison of Pb and Be tracer input, and of Pb and Be mass detected. For yield determination the hydroxides were redissolved in 6 M HCl and Pb and Be determined by ICP-AES. For Cs a single test measurement comparing this method with ion exchange indicates a yield of ~40%.

Results

Lake sediments

The Muttsee sediments are laminated. Millimeter thick light-grey layers that alternate with darker layers of similar thickness are interpreted to indicate seasonal variations caused by differences in the particle input and settling due to ice and snow cover during the cold period. These layers indicate the 'normal' sedimentary lake history. Three events with thickness not exceeding 4 mm record deposition of coarser grains (Fig. 2). A more remarkable event is archived below 15 cm depth, where a homogeneous layer of at least 9 cm thickness forms the deepest part of the

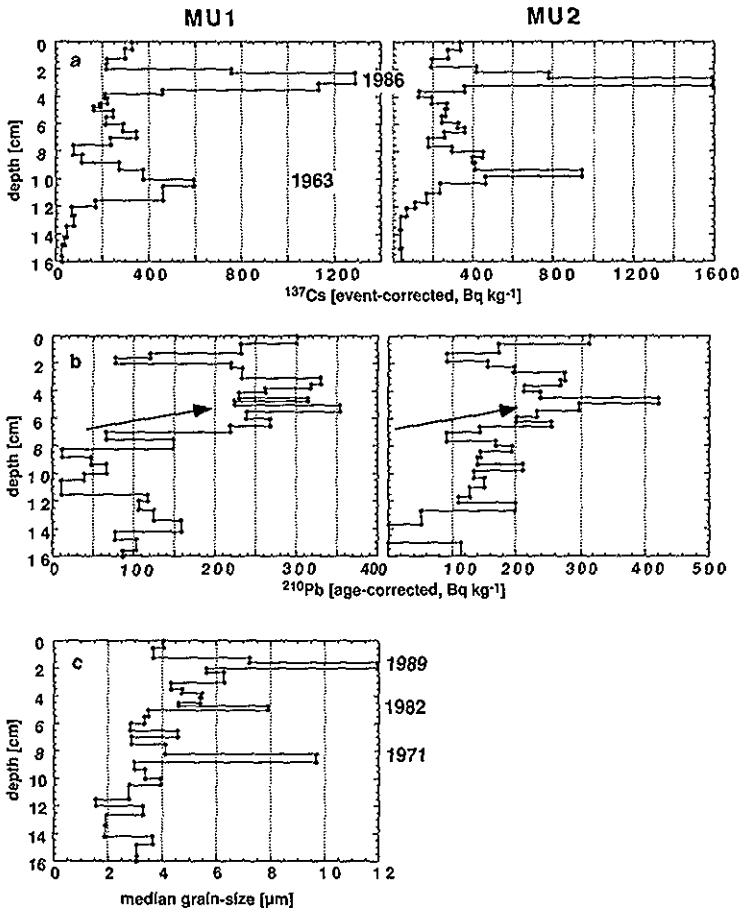

Figure 2. ^{137}Cs (event-corrected, **a**, ^{210}Pb (age-corrected, **b** and median grain-size **c**, as a function of depth in the Muttsee sediment cores 1 and 2. ^{137}Cs allows core dating, ^{137}Cs and ^{210}Pb can be used to indicate radionuclide distribution between catchment and lake and thus inform about the changing soil cover (see text). The arrow (b) indicates the change to increased ^{210}Pb delivery into the lake in the mid 70s. An increase in median grain-size (c) archives a flood event in the catchment area

cores. This 'mixed layer' is likely a result of anthropogenic activity in the lake related to the construction of the pressure duct in the 1950's. This event is of no relevance to our interpretation, which deals only with the younger period.

Organic carbon and total nitrogen were measured in two samples from core 1, one representing a near surface sample (1.2 to 1.6 cm), the second one from an older section (8.2 to 8.8 cm depth). Each sample yielded 0.4–0.5% organic carbon and 0.09–0.1% N. Various lake and river sediments measured in Switzerland (Albrecht, 1995) have significantly higher contents in the range of 2 to 4% for organic carbon and 0.2 to 0.4% for N.

^{137}Cs and ^{210}Pb radionuclide activities and grain-size data are summarized in Table 1. Figure 2a, b shows radionuclide activities for both cores corrected for decay using the time of sediment deposition (^{210}Pb) or time of atmospheric radionuclide deposition (^{137}Cs). The dating of the core is based on the variation of ^{137}Cs with depth, which shows two dominant peaks. The first peak (maxima 590 and 930 Bq kg^{-1} in cores 1 and 2, respectively) represents enhanced deposition as a result of increased atmospheric bomb testing, before the moratorium went into effect at the end of 1963. The second peak, with a maximum of 1300, and 1600 Bq kg^{-1}, respectively, is characterized by the presence of ^{134}Cs (not shown, see Table 1), and reflects the input due to the Chernobyl accident in 1986. For event and age corrections of radionuclide activities (Fig. 2) the average sedimentation rates were calculated based on these time markers, and subsequently ages assigned to all depths assuming constant sedimentation rates between, above and below the two horizons. Radionuclide activities in sediments younger than 1986 are corrected for decay to 1986, those older to 1963, or to the year of deposition for even older samples. The Cs-radionuclide distribution allows determination of sedimentation rates, which increase from 0.3 cm yr^{-1} (average of both cores) during the period 1963–1986 to 0.55 cm yr^{-1} during the period 1986–1992. This calculates to 0.83×10^6 and 1.52×10^6 kg yr^{-1} of dry sediment deposited into the lake. Disregarding sediments, that are related to a flooding event in 1989 (maximum median grain-size; Fig. 2c), still yields a sedimentation rate which is significantly higher (0.45 cm yr^{-1} for core 1) than measured for the earlier period (0.55 cm yr^{-1}). The shift in sedimentation rate is not related to density differences, as seen in the relatively constant water content (42 ± 4 mass-%). Between 1963 and 1986 atmospheric Cs-radionuclide deposition decreased considerably. The deposition caused by continuous atmospheric atomic bomb testing by France in the southern hemisphere had no measurable impact on Switzerland. Chinese tests in the northern hemisphere (concluded in 1981) had a measurable impact on Switzerland but the ^{137}Cs deposition remained several orders of magnitude below the maximum in 1963 (UNSCEAR, 1993; Völkle et al., 1989) and does not need to be considered here. The continued input to the lake, as seen in the sedimentary record, with peaks in the 1960s and 70s (maximum activity = 450 Bq/kg in core 2) therefore indicates delayed and irregular transfer of ^{137}Cs stored in soils from the catchment area to the lake.

The atmospheric input of ^{210}Pb is continuous and has been used to date lake sediments (Krishnaswami et al., 1971). A constant flux of ^{210}Pb into the lake and into the sediments would result in a constant ^{210}Pb activity depth profile. The age-corrected excess ^{210}Pb activities in the sediments of the Muttsee show strong variations (Fig. 2b). Of particular interest is an increase in ^{210}Pb activity in younger sedi-

Table 1. Radionuclide activities in Muttsee sediments

Core 1, 2 sample I.D.	Core sectioning depth [cm]		$^{137}Cs^*$	2σ [%]	$^{134}Cs^*$	2σ [%]	$^{210}Pb^*$	2σ [%]	Particles < 2 μm [%]
	top	bottom							
MU1-1	0.0	0.5	282	16	18	10	297	9	35
MU1-2	0.5	1.2	256	14	15	14	221	10	35
MU1-3	1.2	1.6	187	10	11	23	111	13	21
MU1-4	1.6	2.0	187	10	10	23	69	18	17
MU1-5	2.0	2.3	652	34	46	8	195	10	29
MU1-6	2.3	3.0	1112	61	80	6	201	10	28
MU1-7	3.0	3.5	975	50	69	7	277	10	33
MU1-8	3.5	3.8	394	20	26	13	261	11	30
MU1-9	3.8	4.1	106	6			209	10	29
MU1-10	4.1	4.5	111	6			177	9	31
MU1-11	4.5	4.7	96	7			234	12	33
MU1-12	4.7	5.0	81	5			163	10	26
MU1-13	5.0	5.5	123	7			247	9	38
MU1-14	5.5	6.0	108	6			159	9	39
MU1-15	6.0	6.5	145	9			170	11	42
MU1-16	6.5	7.0	175	10			132	11	32
MU1-17	7.0	7.5	117	7			38	15	43
MU1-18	7.5	8.2	36	3			80	12	33
MU1-19	8.2	8.8	56	4			6	21	23
MU1-20	8.8	9.3	136	8			23	19	42
MU1-21	9.3	10.0	189	10			30	15	38
MU1-22	10.0	10.5	298	16			16	17	35
MU1-23	10.5	11.5	232	13			4	26	44
MU1-24	11.5	12.0	80	6			43	19	56
MU1-25	12.0	12.6	30	2			37	15	40
MU1-26	12.6	13.4	33	2			41	14	51
MU1-27	13.4	14.2	18	2			48	14	52
MU1-28	14.2	14.8	14	1			22	19	36
MU1-29	14.8	15.6	10	1			30	16	39
MU1-30	15.6	16.0	10	1			25	14	38
MU1-31	18.0	18.5	10	1			30	17	36
MU1-32	20.0	20.5	13	2			26	19	34
MU2-1	0.0	0.6	288	6	21	24	329	9	
MU2-2	0.6	1.2	232	6	18	26	187	11	
MU2-3	1.2	1.8	161	6	14	29	105	13	
MU2-4	1.8	2.2	358	5	22	21	160	12	
MU2-5	2.2	2.6	673	6	50	13	193	12	
MU2-6	2.6	3.2	1372	5	98	9	254	11	
MU2-7	3.2	3.6	306	6	19	26	242	10	
MU2-8	3.6	4.0	105	6			190	11	
MU2-9	4.0	4.5	92	7			201	11	
MU2-10	4.5	4.9	128	7			322	10	
MU2-11	4.9	5.4	124	6			222	11	
MU2-12	5.4	5.9	115	7			172	11	
MU2-13	5.9	6.2	154	6			146	12	
MU2-14	6.2	6.6	171	6			174	11	
MU2-15	6.6	7.0	122	6			104	15	
MU2-16	7.0	7.6	84	6			71	14	

Table 1 (continued)

Core 2 sample I.D.	Core sectioning depth [cm]		^{137}Cs*	2σ [%]	^{134}Cs*	2σ [%]	^{210}Pb*	2σ [%]
	top	bottom						
MU2- 17	7.6	8.0	139	6			108	15
MU2- 18	8.0	8.4	215	6			117	14
MU2- 19	8.4	8.8	191	6			90	19
MU2- 20	8.8	9.3	196	6			84	15
MU2- 21	9.3	9.8	448	6			111	16
MU2- 22	9.8	10.3	220	6			75	27
MU2- 23	10.3	11.0	112	7			78	17
MU2- 24	11.0	11.7	38	9			67	28
MU2- 25	11.7	12.1	46	9			58	24
MU2- 26	12.1	12.7	26	11			83	18
MU2- 27	12.7	13.7	14	14			37	41
MU2- 28	13.7	15.0	13	9			18	45
MU2- 29	15.0	16.4	16	8			55	13
MU2- 30	16.4	19.3	15	22			50	41
MU2- 31	19.3	20.7	17	9			61	13
MU2- 32 12. 1992	20.7	22.1	21	7			65	12

* Activities are in Bq kg^{-1} and decay corrected to September 12. 1992, the day of sediment retrieval.

ments. The transition zone between sediments with lower and higher ^{210}Pb activities is at a depth of 6–8 cm. A correlation with grain-size can be observed in certain sections, particularly where a sudden increase in median grain-size occurs. Here ^{210}Pb activities decrease sharply. Sections without a relationship or even a reversed relationship do exist.

Water samples

The radionuclide activities of lake water and surface run-off (Table 2) give evidence of particle retention and radionuclide adsorption. The activities are decay-corrected to the day of sampling (September 2, 1993 and October 13, 1995). Surface run-off that originate in areas partially covered by soil or regolith (sites 2 and 6 in Fig. 1) carry ≤2 Bq m^{-3} ^{137}Cs, <5 Bq m^{-3} ^{210}Pb and <10 Bq m^{-3} ^{7}Be (Table 2 surface run-off from partly soil covered area). Activities from surface run-off interacting only with bare rocks are significantly higher. A surface run-off sample on the S side of the lake had 8.1 Bq m^{-3} ^{137}Cs, 16.2 Bq m^{-3} ^{210}Pb and 52.9 Bq m^{-3} ^{7}Be (Table 2; Site 9). On talus slopes in the NE, E, and SE part of the drainage basin run-off is subterranean. Rocks in these alluvial fans are mostly blocky and platy with low surface area to mass ratios and thus low adsorption efficiency. Drainage through this vadose zone should have radionuclide activities in the same high range as unfiltered surface run-off sampled on the S and N side of the catchment area. The two samples representing lake water are comparable, with activities of ^{137}Cs ranging from 0.45 to 0.68 Bq m^{-3} and ^{210}Pb from 2.2 to 2.5 Bq m^{-3}. The variation in ^{7}Be activi-

Table 2. Radionuclide activities in surface and lake waters

Locality (Figure 1)	sample-ID	sample date	volume [l]	^{7}Be*	2σ **	^{137}Cs*	2σ **	^{210}Pb*	2σ **
surface run-off from partly soil covered area									
SW of lake, (N of 2)	MW I	2.9.93	5	9.5	53	< 0.3		< 4.3	
SW of lake, (SW of 2)	MW VI	13.10.95	25	9.8	13	2.0	14	4.9	18
S of lake, (near 6)	MW III	2.9.93	5	< 1.7		< 0.3		< 1.4	
surface run-off from area lacking soil cover									
N of lake, (E of 5)	MW II	2.9.93	5	46.8	16	4.1	36	11.7	33
S of lake, (at 9)	MW VII	13.10.95	25	52.9	7	8.1	9	16.2	12
lake water									
pressure duct	MW IV	2.9.93	25	15.4	10	0.68	30	2.5	24
pressure duct	MW V	13.10.95	25	6.6	16	0.45	33	2.2	29

* activities [in Bq m^{-3}] are corrected for decay to the day of sampling and for analytical chemical yield (~80% for Be and Pb, 40% for Cs).
** 2σ errors [in %] based on counting statistics.

ties is due to its short half-life and the various storage times within the pressure duct. Because of the short half-life ^{7}Be will not be evaluated further.

Soil and bedrock samples

Soil profiles can be subdivided into 2 categories: (1) those with irregular radionuclide activities (Fig. 3a), and (2) those with regular radionuclide activities with depth (Figs. 3b, c). For all profiles radionuclide activities are decay-corrected to the day of sampling. The activity of $^{210}Pb_{supported}$ can be assessed by measurement of bedrock samples of the three major lithologies; schist, sandstone and sandy marl (Table 3). Activities of ^{210}Pb increase from sandstone (12 Bq kg^{-1}), to schist (23 Bq kg^{-1}), to sandy marl (45 Bq kg^{-1}). A sample of altered schist shows an increase of both ^{137}Cs (75 Bq kg^{-1}) and ^{210}Pb (67 Bq kg^{-1}). This variation is related to the U content of the rock (shale > sandstone), the permeability (sandy marl > schist) and the degree of alteration and thus an increase in surface area per unit mass. The change in rock permeability can best be studied based on the ^{137}Cs activity, which is below the detection level in the schist and the sandstone, but reaches up to 77 Bq kg^{-1} in the sandy marl. The $^{210}Pb_{excess}$ for each soil section is thus based on subtracting the supported ^{210}Pb (the ^{210}Pb of the unaltered underlying bedrock) from the total ^{210}Pb (Table 3). Pb and Cs radionuclides correlate well with depth in individual profiles. In samples from the soil profiles #4 and #5, ^{210}Pb and ^{137}Cs vary between 15 and 140, and 2 and 130 Bq kg^{-1}, respectively. In profiles #3, #6,#7 and #8 ^{210}Pb and ^{137}Cs rise to >5000 and > 900 Bq kg^{-1}. ^{134}Cs is above the detection limit only in samples of category 2, with a maximum of 187 Bq/kg in profile 7.

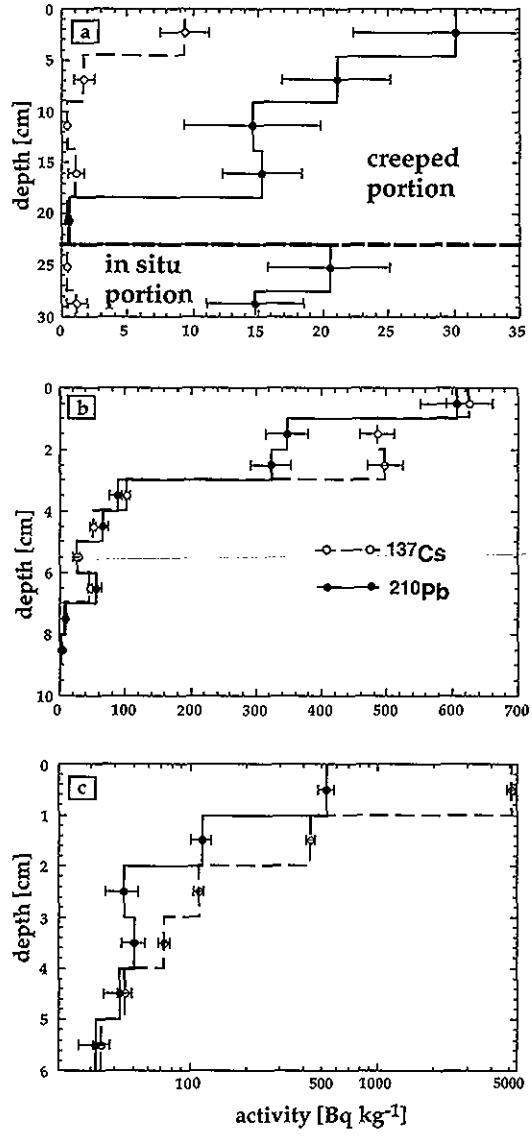

Figure 3. Depth distribution of ^{137}Cs and ^{210}Pb in soil profiles from category 1 (**a**, site 5), where soil creep occurred and stable soils of category 2 (**b**, **c**; sites 3 and 7 – log scale!). The differences in radionuclide activities are related to transient cover (mainly category 1) and to input via unfiltered surface run-off

Table 3. Radionuclide activities in soil and bedrock samples

Depth from	to [cm]	^{137}Cs [Bq kg^{-1}]	2σ	^{134}Cs [Bq kg^{-1}]	2σ	$^{210}Pb_{total}$ [Bq kg^{-1}] see bedrocks	2σ	$^{210}Pb_{excess}$ [Bq kg^{-1}]
category 1, profile #4								
0	1	37	3	–		92	11	70
1	2	102	6	–		130	14	107
2	3	113	7	–		165	18	142
3	4	75	5	–		93	10	71
4	5	100	6	–		93	11	70
5	6	131	8	–		67	11	45
profile #5								
0	5	9.4	1.8	–		42	11	30
5	9	1.7	0.8	–		33	7	21
9	14	≤ 0.5		–		27	10	15
14	18	1.1	0.6	–		27	5	15
18	23	≤ 0.5		–		13	4	1
23	28	≤ 0.5		–		33	7	20
28	30	1.2	0.8	–		27	7	15
category 2, profile #3								
0	1	625	34	21.4	3.7	629	57	606
1	2	485	27	–		370	35	347
2	3	498	27	8.9	2.6	345	33	322
3	4	101	7	–		110	14	88
4	5	51	4	–		87	13	65
5	6	29	3	–		48	10	26
6	7	46	3	–		78	10	55
7	8	9	2	–		32	9	9
8	9	3	1	–		24	7	2
profile #6								
0	1	2389	125	111.4	8.2	894	80	849
1	2	1289	68	66.0	5.7	546	51	501
2	3	690	37	11.1	2.8	338	34	293
3	4	212	12	12.7	2.7	188	20	143
4	5	443	24	10.1	2.5	232	23	187
5	6	8	2	–		40	9	0
profile #7								
0	1	5149	264	187.4	8.6	555	51	532
1	2	437	24	12.9	2.1	137	16	115
2	3	111	7	6.6	2.2	67	13	45
3	4	73	5	4.5	1.4	73	11	50
4	5	45	4	–		65	11	42
5	6	34	3	–		54	11	32
profile #8								
0	1	909	48	16.6	2.6	959	82	937
1	2	384	21	4.8	1.7	307	29	284
2	3	49	3	–		82	11	59
3	4	15	2	–		37	7	14
4	5	8	1	–		29	7	7
5	6	5	1	–		30	7	7
bedrocks								
schist site 1		–		–		23	4	
altered schist site 3		75	5	8.8	2.4	67	10	
sandstone site 7		–		–		12	4	
sandy marl site 6		77	4	12.0	1.9	45	6	

Discussion

A quick status assessment for radionuclide transfer from atmospheric deposition to storage in the lake sediment can be done for the relatively simple Muttsee area on the basis of radionuclide activities in water samples (Table 2). Most particles and radionuclides enter the lake during snow melt in spring and summer. Water is withdrawn from the lake via the pressure duct solely during autumn and winter. The radionuclide levels in lake water in late summer and autumn are significantly lower than those in surface run-off entering the lake (Table 2; pressure duct vs. surface run-off from areas lacking soil cover). This reduction of radioactivity during the summer can only be achieved via particle settling. This efficient reduction of radionuclide levels confirms the results of Santschi et al. (1990), who documented reductions in ^{137}Cs in several Swiss lakes during the summer following the Chernobyl accident in 1986. The small number of surface water samples and the lack of accurate information on monthly precipitation in the Muttsee area, does not allow direct assessment of the residence time of radionuclides in the catchment area solely on the basis of water samples. But water samples clearly show the importance of particle retention and radionuclide adsorption in zones with a soil/regolith cover.

Sediment cores allow estimation of radionuclide and particle fluxes into the lake, determination of bedrock and soil erosion in the catchment area and inventory assessment of radionuclides within the catchment area. As a first step I want to connect sedimentation rates in the lake with erosion rates in the catchment area. The goal is to unravel sedimentation and erosion rates in the last 40 years. Both are related to weathering, and associated climatic and anthropogenic control factors.

The link between sedimentation and physical erosion

The total mass rate of sediment deposited in a lake (M) can be calculated using equation 1:

$$M = \rho_l F_l r_l, \tag{1}$$

where ρ_l is the average bulk sediment density (in g cm^{-3}), F_l the lake surface area (in cm^2), and r_l the mean sedimentation rate (in cm yr^{-1}). The subscripts differentiate lake (l) processes from catchment processes (c). A similar equation allows estimation of masses removed in the catchment area via erosion:

$$M = \rho_c F_c r_c. \tag{2}$$

Here ρ_c indicates the bedrock density, F_c the catchment surface area (excluding the lake) and r_c the erosion rate. The assumption that all eroded material reaches the lake bottom holds for the Muttsee area because of its morphology and because particles have sufficient time for settling before water is withdrawn from the lake via the outlet. Combining equations (1) and (2) yields an expression that relates sedimentation and erosion rates.

$$r_c = \frac{\rho_l F_l}{\rho_c F_c} r_l \tag{3}$$

Table 4. Summary of data pertinent for calculations carried out in the text

(1) general			
approximate precipitation	1800 mm yr^{-1}		
lake surface	0.42 km^2		
drainage area surface (excl. lake)	2.95 km^2		
soil covered area	82 000 m^2		
total volume precipitated	6.1×10^6 m^3		
output volume via duct	5.5×10^6 m^3		
output via evaporation	0.6×10^6 m^3		
(2) densities			used in calculation
bedrock	2.7 g cm^{-3}		
soil	0.4–0.7 g cm^{-3}		0.5 g cm^{-3}
sediment	0.4–0.9 g cm^{-3}		0.66 g cm^{-3}
(3) rates	core 1	core 2	
sedimentation (1963–1986)	0.31	0.29	cm yr^{-1}
sedimentation (1986–1992)	0.57	0.53	cm yr^{-1}
erosion (1963–1986)	10.8	10.1	cm kyr^{-1}
erosion (1986–1992)	19.8	18.4	cm kyr^{-1}
mass removed (1963–1986)	2.9×10^5	2.7×10^5	kg yr^{-1} km^{-2}
mass removed (1986–1992)	5.3×10^5	5.0×10^5	kg yr^{-1} km^{-2}
(4) radionuclide deposition			
^{137}Cs atmospheric atomic bombs	5.5 KBq m^{-2}		(Wan et al., 1987)
	3.6 KBq m^{-2}		(this paper)
^{137}Cs Chernobyl	2.3 KBq m^{-2}		(KUeR, 1989)
^{210}Pb	140 Bq m^{-2} yr^{-1}		(Schuler et al., 1991)
(5) Budgets	core 1	core 2	
^{137}Cs Chernobyl, total deposition	7.8×10^9	7.8×10^9	Bq
^{137}Cs Chernobyl, in sediments	6.0×10^9	6.9×10^9	Bq
	or 77.6	or 88.7	%
^{134}Cs Chernobyl, total deposition	4.4×10^9	4.4×10^9	Bq
^{134}Cs Chernobyl, in sediments	3.0×10^9	3.6×10^9	3.3×10^9 Bq
	or 69.4	or 83.5	76.4 %
^{210}Pb, total deposition 1986–1992	3.0×10^9	3.0×10^9	Bq
^{210}Pb, in sediments	2.2×10^9	2.6×10^9	2.4×10^9 Bq
	or 74.4	or 84.8	79.6 %
^{137}Cs atmospheric atomic bomb deposition			
sediments younger 1986	3.1×10^9 Bq	26 %	
sediments older 1986	7.6×10^9 Bq	63 %	
soils	1.4×10^9 Bq	12 %	
total	12.1×10^9 Bq	**thus 3.6 KBq m^{-2}**	

Density remains relatively stable as a function of depth but sedimentation rates have changed as archived by the 2 sediment cores. Rates are therefore not represented as differentials, but as averages over well defined time or sampling periods, e.g. 1963–1986 or 1986–1992. Based on information summarized in Table 4 it is possible to calculate erosion rates averaged over the catchment area. The sedimentation rates of 0.31 and 0.57 cm yr^{-1} for the periods 1963–1986 and 1986–1992 yield erosion rates between 10.5 and 19.1 cm kyr^{-1}. These rates calculate to 2.8×10^5 and 5.2×10^5 Kg km^{-2} yr^{-1}, respectively. This is rapid, when compared with erosion rates on stable surfaces such as a rhyolite plateau in the arid climate of New Mexico, where maximum model erosion rates, based on cosmogenic nuclides, do not exceed

1.1 cm kyr^{-1} (Albrecht et al., 1993). Regional rates, given in Kg km^{-2} yr^{-1} vary between an average 2.7×10^4 for Europe, 5×10^4 for the lower Lake Biel catchment, (Santschi and Schindler, 1977), 9.5×10^4 for Lake Sempach and 4.5×10^4 for Lake Zürich (Schuler et al., 1991; Wieland et al, 1993; all three in the Swiss lowland) and $\sim 1 \times 10^6$ for the catchment areas of Lake Geneva and Lake Constance (Swiss alpine region, Li and Erni, 1974). The value for physical erosion $\sim 5 \times 10^5$ Kg km^{-2} yr^{-1} for the Muttsee area represents an intermediate number. Comparison of local with regional erosion rates is a little risky, because the methods rely on different assumptions. In our case the erosion rate is based on a homogeneous sediment deposition across the entire lake bottom, which was evaluated on the basis of only two sediment cores, while other studies used cosmogenic radionuclide activities in exposed quartz or sediment fluxes in rivers and lakes. Border zones of lakes, particularly steep walls, tend to have lower sedimentation rates. Subtracting these areas from F_1 would bring a reduction of erosion rates by ~ 10 %. It is nonetheless promising that differences between our and previous estimates can well be explained by natural variation.

A striking result derived from the radionuclide depth profiles is the recent increase in the mean erosion rate. The depth distribution of both ^{137}Cs and ^{210}Pb and the varying amplitudes of the 1963 ^{137}Cs peaks (Figs. 2a, b) compared to the 1986 maximum indicate that both particles and radionuclides were held back in the catchment area more efficiently in the earlier period than during more recent times. A possible explanation would be a greater regolith or soil cover during the early sixties and a subsequent removal of parts of this cover. Atomic bomb related ^{137}Cs deposition in the catchment area became insignificant at the end of 1963, but input into lake sediments continued. Major flood events are archived in the sediment core and can be found using the grain-size depth distribution (Fig. 2c). The post-Chernobyl flood event (~1989) could flush the catchment area efficiently because the soil cover was probably as patchy as it is today. The post-bomb flood event (~1971) could not as efficiently remove the bomb ^{137}Cs, because of a more extensive, erosion resistant soil cover. The additional maxima in both cores (Fig. 2a), dated to approximately the mid-sixties and mid-seventies can be interpreted as the removal of sections of top soil with high radionuclide content.

The significant increase in ^{210}Pb activity (see arrows in Fig. 2b) from <200 to >200 Bq/kg can be explained by increased delivery of particle bound ^{210}Pb from the catchment to the lake. It coincides with the mid-1970s ^{137}Cs peak and gives therefore further evidence for a change in soil/regolith cover and erosion regime. This episode is not related to a major flooding event as no significant sediment grain-size jump can be observed in the transition zone. The soil erosion must have been related to a considerable extent to a landslide that occurred mostly on the foot of the Nüschenstock (Fig. 1) in the early sixties, covering areas previously supporting grass and soil (H. Caduff, personal communication). Some of this disturbed section was eroded and transported into the lake shortly after 1963, further removal occurred during the mid-seventies, as can be seen in the radionuclide depth profiles. The difference between core 1 and 2 finds the origin also in this landslide. As core 2 is located more closely to the foot of Nüschenstock (Fig. 1), it archived this local event and the related delivery of particle bound radionuclides more effectively.

Radionuclide budget and evaluation of radioactivity in lake sediments

The present distribution of soil/regolith and lake sediment radioactivity can be evaluated using a mass balance approach. The total deposition of radionuclides into the lake and the catchment area is given by multiplication of the atmospheric deposition (q, for ^{210}Pb as a flux per year and for ^{137}Cs per event) with the lake and catchment surface area (F_l, F_c). All compartments that play a role in an inventory formula $I_{tot} = I_l + I_c + O$ can be sampled. Output (O) is via the pressure duct. With an annual volume of 5.5×10^6 m^3 and a ^{210}Pb and ^{137}Cs activity of 2.35 and 0.57 Bq m^{-3} (average of 2 samples; Table 2), a total of 1.3×10^7 and 3.1×10^6 Bq, respectively leave the Muttsee watershed per year. The possibility of higher ^{137}Cs output via the duct during the winter 1986/87 can not be ruled out, but the extent should be relatively small due to efficient scavenging during spring/summer 1986. The remainder, which is more than 99%, resides in lake sediments (I_l) or is bound to soil or regolith particles within the catchment (I_c). Inventory calculations for lake sediments are performed for both cores (Table 4) based on event corrected ^{137}Cs- and age-corrected ^{210}Pb activities (Fig. 2a, b). The time of deposition of each individual sample is based on the sedimentation rates determined for each core for the periods 1963–1986 and 1986–1992. The inventory of lake sediments can be assessed using the following equation:

$$I_l = \left(\sum_{i=1}^{n} a_i \rho_i \Delta t_i \right) r_l F_l \qquad (4)$$

where a_i, ρ_i and Δt_i are activity, bulk dry density and deposition time of each individual sample in the sediment archive (with n the last sample, Table 4). For ^{137}Cs, an inventory (I_l) is calculated for the periods 1963–1986 and 1986–1992, for ^{210}Pb only for the more recent period. An additional calculation for ^{134}Cs was performed for the period 1986–1992. The continuous input of ^{210}Pb makes age corrections in older sediments ambiguous, because radionuclides may have been deposited directly into the lake with subsequent incorporation into the sediment, or may have been transported into the lake adsorbed to particles with various catchment area residence times. For ^{137}Cs this problem only pertains to the portion of radionuclides that reached the catchment area during 1963, but remained there past 1986. The total deposition for atomic bomb related ^{137}Cs based on Wan et al. (1987) calculates to be 18.5×10^9 Bq, with only 7.6×10^9 Bq deposited in sediments up to 1986 (=41%). As noted before, the deposition rate for the atmospheric atomic bomb tests is unreliable. The decrease in ^{137}Cs delivery to the lake in the 1970's and early 1980's and the sharp peak of 1986 above a flat baseline (Fig. 2a) give evidence of more efficient removal of bomb ^{137}Cs by 1986. The radiocesium budget for Chernobyl allows a reevaluation of the ^{137}Cs atomic bomb deposition. The total deposition for the Chernobyl event is much better constrained (KUeR, 1989; Santschi et al., 1990) and amounts to 7.8×10^9 Bq for ^{137}Cs and 4.3×10^9 Bq for ^{134}Cs. For ^{134}Cs, 3.3×10^9 Bq are found in lake sediments, thus 76% of the total. The same portion of sediment bound activity for Chernobyl-related ^{137}Cs yields 5.93×10^9 Bq. As the total activity in sediments amounts to 7.76×10^9 Bq, the difference of 1.83×10^9 Bq has to have its origin in bomb deposition. Decay corrected to 1963 yields

3.11×10^9 Bq, which has to be added to the bomb-related ^{137}Cs bound in sediments older than 1986 (Table 4). This budget for bomb radiocesium will be completed on the basis of the soil radioactivity.

A ^{137}Cs inventory study, carried out in northern Sweden (Malmgren and Jansson, 1995) found 10% of the total deposition to be removed from the catchment and 90% of this portion was transported to the outer sea of the Gulf of Bothnia. ^{137}Cs was interpreted to be mostly dissolved or in colloidal form, possibly associated with dissolved organic material. At Muttsee the organic material is insignificant as seen by low concentrations of organic carbon and nitrogen in the sediment (0.5 and 0.1%, respectively). Adsorption to clays such as illite (Comans et al., 1991; Comans and Hockley, 1992) is thus likely the controlling process explaining the efficient transfer of radionuclides into the sediment.

The budget for ^{210}Pb is calculated for the period May 1986 to September 1992 (=6.4 years). Total calculated deposition amounted to 3.0×10^9 Bq, of which 2.4×10^9 Bq or 80% are found in lake sediments. The similarity between the results obtained for Chernobyl ^{134}Cs (76%) and natural ^{210}Pb (80%) is most encouraging. Furthermore these values coincide with the relative surface area of the lake (84%) compared with the total radionuclide storage area (lake + soil covered area, Table 4). As the critical atmospheric input functions of ^{137}Cs and ^{210}Pb are based on different data sets, the similarity in the sediment inventory for both radionuclides reflects on the quality of choice for these input terms. The remaining ^{210}Pb and radiocesium can be found in the soil and regolith cover of the catchment area.

Radionuclides in the soil component

The soil profiles taken from 6 different localities show considerable variation both in regard to ^{210}Pb and radiocesium (Table 3; Fig. 3). ^{137}Cs maxima in individual soil samples vary from 9 to 5000 Bq kg^{-1} (for a person who spends one hour per day on such a soil the maximum activity calculates to a dose of 0.2 mSv yr^{-1}, 20% of the acceptable limit). The large range of activities makes an assessment of higher activities at more strongly contaminated sites quite difficult. One possibility to ensure that the range sampled is indeed representative is to increase the number of samples or to take advantage of portable γ-spectrometers (Murith et al., 1986).

In the case of the Muttsee catchment we can use the results obtained from the radionuclide budget to evaluate the representativeness of our soil samples. About 76% of the total inventory of ^{134}Cs (and as a consequence of comparable chemical behaviour also ^{137}Cs) was found in lake sediments (Table 4). As the portion of ^{137}Cs that left the system through the pressure duct is less than 1%, the remainder, or 1.8×10^9 Bq of ^{137}Cs and 1.0×10^9 Bq of ^{134}Cs, has to reside in the soils of the catchment. Distribution of this radioactivity in the upper 2 cm of soils covering 82000 m^2 calculates to a model soil activity for ^{137}Cs and ^{134}Cs of 2230 and 1239 Bq kg^{-1}, respectively (during deposition in May 1986, using the average measured soil density of 0.5 g cm^{-3}). As this is well within the range of soil actually sampled, our samples can be classified as representative and no doses higher than the one obtained from the maximum soil activity would be expected within the catchment area.

We can perform an additional computation on the basis of these soil samples. The event corrected $^{137}Cs/^{134}Cs$ ratio of 2.0 in post-Chernobyl sediments is slightly elevated compared to the ratio of 1.8 measured during the event. The transfer of bomb-related ^{137}Cs from the catchment to the sediment after the Chernobyl event has been evaluated using these ratios. Similar calculations allow an estimate of the fraction of bomb-related ^{137}Cs that still resides in catchment soils. Measured $^{137}Cs/^{134}Cs$ ratios in soils are as high as 6.2, with an average of 2.6. In soils 24 % of the total Chernobyl radiocesium reside with a ratio of 1.8. It requires an additional 0.82×10^9 Bq of bomb-related ^{137}Cs to raise the ratio to 2.6. This activity corrected for decay to 1963 (1.4×10^9 Bq) has to be added to the bomb-related ^{137}Cs found in sediments (Table 4). The total deposition in the catchment and lake area due to atomic bomb testing thus adds up to 12.1×10^9 Bq, of which 88 % now reside in lake sediments and 12 % in catchment soils. The total bomb related deposition per area equals 3.6 kBq m^{-2}. This is a more accurate determination than the previously published estimate of 5.5 kBq m^{-2} (Wan et al., 1987).

Radionuclides as a pedologic tool

Soil formation and denudation processes form important links to the understanding of weathering and erosion. Radionuclides such as ^{10}Be, ^{210}Pb ^{137}Cs and have been used to quantify soil ages, trace eroded soils and evaluate soil erosion, estimate bedrock-to-soil conversion rates and to recognize paleosols (Pavich et al., 1985; Monaghan et al., 1992; Wallbrink and Murray, 1993; Middleton et al., 1994). An attempt will be made here to evaluate ^{210}Pb plus radiocesium for soil movement description.

The increase in radionuclide activity from bedrock to regolith to soil (sites 1, 3; Fig. 1; Table 3) is a consequence of the larger number of surface sites available for adsorption per unit mass (Stumm and Morgan, 1996) resulting from physical weathering. An unaltered schist shows no measurable activity of ^{137}Cs; for ^{210}Pb a value of 23 Bq kg^{-1} was measured. In a sample of altered 'muddy' clay ^{137}Cs and ^{210}Pb increase to 75 and 67 Bq kg^{-1} and in the top soil layer both exceed 600 Bq kg^{-1}. The strong variation from soil profile to soil profile (Fig. 3) is not caused by differences in adsorption efficiency but is related to different input functions. As the soil cover in the catchment area is patchy, the soil receives radionuclides not only directly from the atmosphere, but in addition laterally from non-filtered surface run-off. A closer inspection of depth profiles points toward a qualitative approach, which may indicate an additional pathway for soil morphologists. In soil profile #4 the ^{210}Pb activity first increases then drops again (Table 3). In profile #5, ^{210}Pb drops close to the zero level and then increases again with depth (Fig. 3a), allowing the conclusion that the upper section in these profiles dit not form in situ, but must have creeped or slid in and covered the lower soil portion. Low activities and the absence of ^{134}Cs indicate a substantial cover of these soil sections during much of the Chernobyl event with subsequent removal of the transient cover. Creep, slide and cover phenomena have been found only in soil profiles that are located near Nüschenstock, suggesting a relation to the landslide that occurred in the early 60s. Maximum ^{137}Cs concentrations in profiles which were not periodically covered indicate the surface layer during the Chernobyl accident.

Summary

1. The overall budget of radionuclides stored in each compartment of a small alpine catchment area is consistent. The samples are therefore representative for the actual radioactivity present in the catchment. About 80% of ^{210}Pb and $^{137,134}Cs$ deposited are stored in sediments, the remaining portion remain in the soil cover. These fractions compare well with the individual fractions of the total radionuclide storage area, 84% for the lake and 16% for the soil-covered area. Probably less than 1% left the system via aquatic transport through a man-made pressure duct. Intercompartment transfer of radionuclides is mostly in particulate form.
2. The results indicate the applicability of radionuclides to study high mountain exogenic dynamic processes. Sedimentation rates in the lake can be determined on the basis of the ^{137}Cs depth distribution and range from 0.3 to 0.6 cm yr^{-1}. This corresponds to erosion rates varying between 10 and 20 cm kyr^{-1}. Sedimentation and erosion rates, during the most recent period, have increased compared to the period 1963–1986.
3. On the basis of radionuclide budget and radionuclide depth profiles it can be concluded that the soil and regolith cover of the catchment was more extensive before the early sixties compared to the present cover. The change in sedimentation and erosion rate and extent of soil cover is mostly related to a landslide that took place during the early sixties.
4. In today's patchy, thin soil cover ^{210}Pb input is both via direct precipitation and via unfiltered surface run-off, giving rise to strong variation in soil radionuclide activities.
5. Radionuclides can be used as a pedologic tool by analysing soil depth profiles. They allow recognition of soil movement or transient cover of newly-exposed soil sections, such as those associated with the landslide in the early 60s.

Concluding remarks

The reader might be compelled to compare this study with similar attempts done in different settings. Malmgren and Jansson (1995) estimated that 10% of ^{137}Cs inputs were bound to lake and estuarine sediments, quite different from 80% estimated for the Muttsee catchment. Brittain et al. (1992) found a higher extent of ^{137}Cs removal by sediments. These differences are not surprising in view of varying metal behaviour as a function of river chemistry and suspended particle mineralogy, both factors still awaiting a consensus regarding understanding and quantification.

The study was intended as an example to evaluate the radionuclide behaviour in a specified field area, and developed into a study that linked geomorphic approaches of erosion rate determination and radionuclide dating of lake sediments with soil dynamics. Expecting considerable complexity, I chose a most simple alpine lake region, characterized by mostly barren rocks, void of major biologic and anthropogenic influence. Moving down to lower elevations will increase the catchment surface and thus the geologic, biologic and hydrologic complexity. The key to test at least regional models of understanding has to be the search for the 'simplest' natural study sites, such as the Muttsee area and to uncover hidden links between compartments using multidisciplinary approaches. This will allow to make

small steps toward a more reliable and less ambiguous quantified understanding of natural catchment processes.

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