

Importance of lacustrine physical factors for the distribution of anthropogenic ^{60}Co in Lake Biel

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Abstract

The fate of particle reactive substances in a surface aquatic system is controlled by both geochemical and physical parameters. Scavenging processes in Lake Biel (Switzerland) have been studied using the anthropogenic tracer ^{60}Co . This isotope originates mostly in dissolved form from the Mühleberg nuclear power plant and is transported 18 km down by the River Aare to the lake. Radionuclide data from sediments, combined with inventory calculations, indicate that between 30% and 55% of the discharged ^{60}Co is transferred to sediments. Scavenging efficiency is higher during the winter period than during summer, in spite of higher particle fluxes during the summer. An intrusion model, based on buoyancy calculations, predicts that Aare water, including its radionuclide load, remains in the epilimnion during the warm period; thus, mixing is restricted to only a fraction of the lake volume and the residence time is reduced to 1 week. Buoyancy in the summer is controlled mostly by temperature. During winter, when the lake and Aare waters are close to 4°C, the higher dissolved load of Aare water increases its density and forces it to intrude into deeper sections of the lake, increasing the residence time and thus the time available for adsorption and particle settling. The predictive capability of the model was tested for the summer period using a fluorescent dye (uranin) to track Aare water from the inflow to the outflow of the lake. Higher fluxes of both particles and associated radionuclides in deep sediment traps confirm the results of the intrusion model for the winter period. Comparison between modeled and measured ^{60}Co activities in lake water at the end of the winter validates the credibility of the intrusion model further. This combined geochemical and physical approach may be applied to transport modeling of radionuclides and of any particle reactive substance, inorganic or organic.

In general, dissolved metals (e.g., radionuclides) are removed from aquatic systems mainly by adsorption onto particles and subsequent sedimentation. In the past, understanding of metal adsorption has been shaped by thermodynamic principles. The time scale of the adsorption process was considered of minor importance because reactions were assumed to happen quickly (Buffle 1988; Buffle and van Leeuwen 1992; Stumm and Morgan 1996; Garnier et al. 1997). However, for systems with high particle to water ratios (e.g., soils), equilibrium is not an appropriate representation of field conditions. Therefore, kinetic models are needed to understand metal mobility with time (Sparks 1995). The shorter residence times of substances in surface freshwater systems emphasize the importance of the time scales of chemical reactions in such systems. Stoke's law predicts that the second step in metal removal, particle settling, is fast for larger particles (up to few meters per second), but for the most important particles, i.e., clays, oxides, and colloidal aggregates

of $<2 \mu\text{m}$, settling velocities are significantly smaller (meters per hour or per day, O'Melia and Bowman 1984; Weilenmann et al. 1989). These two time factors, the contact time between sorbate and sorbent and the residence time, significantly influence metal behavior in surface aquatic systems and are both controlled by aquatic physics.

Lake Biel, a natural lake system located in northwestern central Switzerland (Fig. 1), is an almost ideal field site to demonstrate the importance of aquatic physics to the transport of radioactive metals, either dissolved in water or adsorbed onto particles. Regulated amounts of anthropogenic radionuclides are released on a regular basis into the River Aare by the Mühleberg nuclear power plant, located 18 km upstream from Lake Biel. These discharges represent the only source of the radionuclides ^{60}Co , ^{54}Mn , and ^{65}Zn within the study area. The focus of our study is on ^{60}Co because it is the principle radionuclide in the waste water and because it has the longest half-life (1,925.6 d compared with 312.2 d and 243.8 d for ^{54}Mn and ^{65}Zn , respectively). Average water residence times of approximately 60 d in Lake Biel are related to the high runoff of the River Aare. This unnatural predominance of the River Aare is also expressed in the rate and the type of sedimentation within the lake (Weiss 1979; Wright et al. 1980). In the Aare delta, sedimentation rates are as high as 3.7 cm yr^{-1} but drop in the deeper basins to $<2 \text{ cm yr}^{-1}$ (Albrecht et al. 1997). These dated sediments allow evaluation of both temporal and spatial distributions of radionuclides, a comparison with discharges by the nu-

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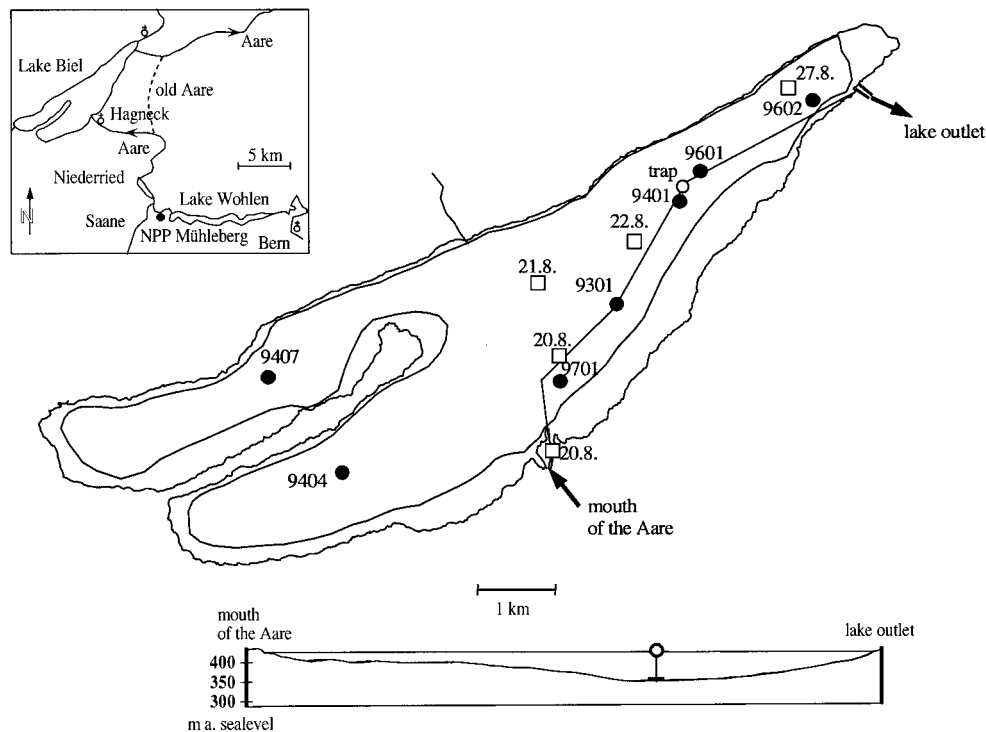


Fig. 1. Map of Lake Biel with all sampling sites (● = sediment cores; □ = sampling sites for uranium profiling; ○ = the sediment mooring site at the deepest location of the lake), the zone of stable sedimentation (white), and the littoral zone (gray). The upper insert shows the lower catchment and the location of the Mühleberg nuclear power plant along the Aare River. The lake cross-section in the lower part of the figure indicates the lake morphology along the black line shown on the lake map.

clear power plant, and the formulation of a sedimentological model. To test the conceptual sediment deposition model and to link geochemical and physical processes, a detailed investigation of the intrusion of Aare water into the lake has been carried out. The results of this year-long combined investigation of sediment deposition and lake physics are presented here. Special emphasis is placed on radionuclides in trap sediments and water samples as a function of water depth and season and on the use of natural tracers such as temperature and artificial tracers such as fluorescent dyes to locate river water within the lake.

Lake Biel—Lake Biel is an atypical alpine lowland lake strongly dominated by the Aare River, which flows into the lake as a result of manmade channels built in the last part

of the 19th century (Fig. 1). The large water discharge of the Aare relative to the lake volume results in a water residence time of ~ 60 d, which is very short compared with the 13-yr residence time for Lake Constance and Lake Geneva.

Further hydrological and morphological properties of Lake Biel are summarized in Table 1. The water of Lake Biel is eutrophic and hard, with total PO_4^{3-} close to $100 \mu\text{g liter}^{-1}$ in the 1970s, dropping to $50 \mu\text{g liter}^{-1}$ in the early 1990s, and a Ca content of 60 mg liter^{-1} (Wright et al. 1980).

Based on published information (Weiss 1979; Wright et al. 1980), we have subdivided the lake into a zone of relatively undisturbed, continuous sedimentation and a shallow water zone, where sedimentation and erosion rates are comparable (Fig. 1). The zone of continuous sedimentation occupies a total area of 30.2 km^2 (Table 1). The dominance of Aare suspended particles is most obvious in the tongue-shaped Aare delta. They can be traced using the clay or carbonate content of the surface sediments. After entering the lake, the Aare drifts off from a northern to a northeastern direction, moving preferentially to the lake exit (Weiss 1979). Current measurements suggest that the right-lateral entrainment of Aare water causes a preferred deposition of Aare particles during high water events and a distinct turbid layer in the NE basin. In addition it induces general counterclockwise water circulation in the lake at all depths (Nydegger 1976a; Wright and Nydegger 1980).

Distinct phytoplankton blooms develop during spring and midsummer, with similar primary production in all three ba-

Table 1. General information on Lake Biel.

Character	Value
Average altitude of lake level (m above sea level)	429
Total surface area (km^2)	39.3
Surface area of net sedimentation (km^2)	30.2
Length (km)	15
Maximum width (km)	4.1
Maximum depth (m)	74
Average depth (m)	30.5
Volume (km^3)	1.18
Average discharge (1935–1990, $\text{m}^3 \text{ s}^{-1}$)	244

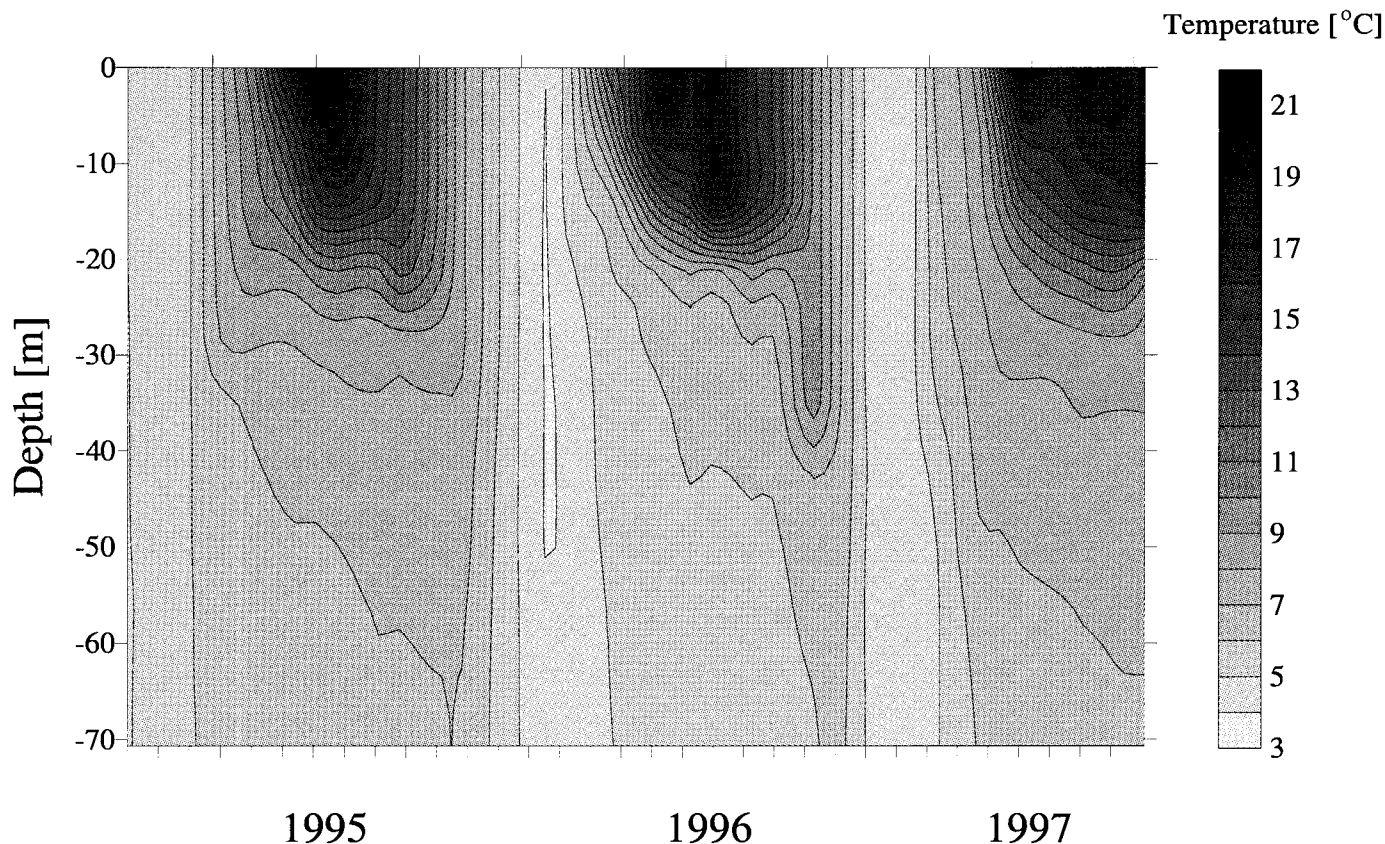


Fig. 2. Isotemperature plot from 1995 to 1997 based on temperature profiles measured at the deepest location of the lake basin (trap, Fig. 1). As in most lakes, temperature increases in the upper layers during the warm period (mid-April to mid-October), and a stable density stratification develops. During the coldest period, the lake is completely mixed.

sins (Tschumi 1977). Anoxia occurs only between September and November in the deepest basin (NE basin) after peak primary production.

Physical characteristics are similar to those of other Swiss lowland lakes. During the warm period (mid-April to mid-October), temperature increases in the upper layers and a stable density stratification develops. The depth of the thermocline varies throughout the year as a function of heat flux and wind forcing (Fig. 2). Because of the low solute content (0.22–0.32‰), the density of lake water is mainly controlled by temperature. In contrast to this, solute load in the River Aare (0.25–0.65‰) has a considerable impact on its density. In particular during winter, this contribution must be taken into account for the computation of the intrusion depth.

Radionuclide source and transport to the lake—Artificial radionuclides (mostly ^{60}Co , ^{65}Zn , ^{54}Mn , and ^{137}Cs) originate from the Mühleberg nuclear reactor, a boiling-water-type nuclear reactor located on the Aare River, about 13 km WNW of the Swiss capital of Bern (Fig. 1). Discharges of these radionuclides are known and occur on an irregular weekly to biweekly basis. ^{60}Co , the predominant radionuclide, has a half-life of 5.3 yr and is produced by neutron activation from stable ^{59}Co , used in high resistivity steels. Total monthly discharges for the period January 1991–May 1997 show maxima during summers (about 10^9 Bq month $^{-1}$), which are

related to reactor revisions. Increased discharges, like those during August 1982 and the winter months of 1993/1994, were related to exceptional torus cleaning efforts.

Radionuclide activities and chemical forms in the wastewater before release vary among discharges. Filtration with a particle size cutoff of 0.2 μm and ion-exchange has shown that the major portion of ^{60}Co is “dissolved” and in cationic form. After discharge at the nuclear installation and a travel time of 8–15 h for the 18.8-km downstream distance (depending on river runoff), the radionuclides arrive at Lake Biel. River sediment analysis and comparison of modeled and measured breakthrough curves along the course to the lake indicate that >95% of the discharged ^{60}Co reaches the lake, of which <20% is adsorbed to particles >0.5 μm . The particle load in the river (2–160 g m $^{-3}$) varied mostly as a function of runoff of the Aare tributary Saane (Fig. 1). Particle composition is dominated by sheet silicates and organic material (50%), with additions of carbonates (25%), quartz (15%), and feldspars (10%) (Albrecht et al. 1995).

During a coordinated discharge in September 1993 carried out to investigate the river section between the reactor and the lake, the first measurements of the ^{60}Co activity were carried out in Lake Biel (Albrecht unpubl. data). At Hag-neck, a maximum activity of 20 Bq m $^{-3}$ was simulated with a mathematical river model (AQUASIM, Reichert, 1994), and activities of 17 ± 1.8 and 11 ± 1.1 Bq m $^{-3}$ were mea-

sured. Three samples were taken within the delta at a distance of approximately 700 m from the mouth of the Aare. At the surface, mean (\pm SE) activity was $0.12 \pm 0.02 \text{ Bq m}^{-3}$, which increased with depth to $0.23 \pm 0.03 \text{ Bq m}^{-3}$ at 10 m and to $1.7 \pm 0.3 \text{ Bq m}^{-3}$ at 15 m depth. The same day, an activity of $0.09 \pm 0.02 \text{ Bq m}^{-3}$ was measured at the outlet of the lake near Biel, 9 km from the Aare inlet (Fig. 1). Five days later the activity of ^{60}Co increased to $3.9 \pm 1.9 \text{ Bq m}^{-3}$, as a result of the discharge carried out 6 days earlier.

Methods

Radionuclides in the water column—To determine the activity of radionuclides in the water column, 100- and 200-liter water samples were taken near the deepest location of the lake (site *trap*, Fig. 1). Radionuclides were concentrated by precipitating $\text{Fe}(\text{OH})_3$ (pH 8) and freeze-drying the precipitate before γ -spectroscopy. Precipitation yield (30–50% for Co) was determined based on stable metal addition during sampling and inductively coupled plasma-atomic emission spectroscopy (ICP-AES) determination after redissolution (Albrecht et al. 1995).

Radionuclides in the sediment—Sediment cores were taken at different locations in the three basins, within the Aare delta and near the lake exit (Fig. 1). Cores were sampled at 5–20-mm intervals on site and freeze-dried before γ -spectroscopy. Dating of sediments relies primarily on the two distinct ^{137}Cs time markers; the first related to atmospheric deposition during the early 1960s (nuclear test peak) and the second in 1986 (Chernobyl accident). Fluctuations of the ^{137}Cs and ^{60}Co discharges by the nuclear reactor and the release of anionic surfactants can be seen in the sediment record and allow independent confirmation of sedimentation rates (Albrecht et al. 1998; Reiser et al. 1997).

Radionuclides in trap sediments—A set of three cylinder-shaped sediment traps were installed between 3 July 1996 and 4 April 1997 at the deepest location of the northern lake basin (*trap* in Fig. 1). Each trap was equipped with two separate cylinders, each with a collecting surface area of 66.5 cm^2 . The first trap was positioned just below the epilimnion (22 m below the lake surface), the second within the hypolimnion (46 m below the surface), and the third 2 m above the bottom (72 m below the surface). Samples were collected at irregular intervals, ranging from 2 d to 2 months. Dry weight was determined after freeze-drying, and the radioactivity was measured by γ -spectroscopy.

Fluorescence, conductivity, and temperature profiles—For experiments in lakes, the fluorescent dye uranin (sodium fluorescein) is an appropriate optical tracer with high fluorescence efficiency, low absorbance to particulate and organic matter, and no known toxicity to aquatic organisms (Smart 1982). Fluorescence profiles were measured at a sampling rate of 10 Hz, using an in situ fluorometer (Dr. Haardt, Kleinbarkau, Germany). This instrument had a large detection range of $0.01\text{--}700 \text{ mg m}^{-3}$ for uranin concentration, was calibrated in the laboratory, and was attached to a commercial conductivity-temperature-depth (CTD) probe (Sea Bird

Electronics). This equipment allowed the simultaneous measurement of depth, temperature, salinity, and fluorescence. The ship's position was determined by a differential global positioning system (Garmin) with a resolution of approximately 10 m. In the Aare River, uranin concentrations were measured with a second in situ fluorometer (LOT) with a smaller but adequate detection range ($0.2\text{--}200 \text{ mg m}^{-3}$ uranin) and sampling rate (4 Hz). Additionally, CTD profiles were regularly recorded every month at the deepest point of the lake (site *trap*, Fig. 1). These measurements allow precise monitoring of the annual development of the thermocline (Fig. 2).

Auxiliary methods—Particle concentration was determined using different methods: classical filtration using $0.45\text{-}\mu\text{m}$ nucleopore filters (50 mm diameter), in situ filtration using $0.45\text{-}\mu\text{m}$ Gelman Supor[®] -450 filters (293 mm diameter), and continuous flow centrifugation at 30,000 rpm, with an approximate particle cutoff at $0.5 \mu\text{m}$.

Particle mineralogy was studied by X-ray diffraction, using the SCINTAG XDS 2000 at the University of Neuchâtel. Grain-size spectra were analyzed using a laser-equipped Malvern Mastersizer X particle sizer (lens with a 100-mm focus). Dried samples were resuspended in water and analyzed after 1 min of ultrasound treatment.

Results and Discussion

Radionuclide inventory and distribution—The prerequisites for carrying out an inventory study in Lake Biel are (1) known input of the substance of interest (in this case ^{60}Co), (2) dated sediment archives, and (3) sufficient sediment homogeneity to allow interpolation from a few cores to the entire lake. Seven sediment cores were taken in Lake Biel to test the feasibility and possible errors of such an inventory calculation. The cores were taken to represent the most important zones of sedimentation within the lake (Wright et al. 1980). In addition to nuclear-test-related and Chernobyl-related ^{137}Cs , higher discharges by the Mühleberg nuclear reactor resulted in two additional markers, one of ^{137}Cs and the second of ^{60}Co at depths representing 1977/78 and 1982, respectively. The four peaks enable more accurate dating in Lake Biel than in other lakes (Albrecht et al. 1998; Fig. 3). Based on seven sediment cores, the spatial variation in the sedimentation rate varies from a maximum of 3.7 cm yr^{-1} in the Aare delta to a minimum of $<2 \text{ cm yr}^{-1}$ in the deeper basins (Albrecht et al. 1997, 1998).

Mass-related ^{60}Co activities (A) (Bq kg^{-1} dry sediment) are first corrected for decay to the date of deposition. Conversion to fluxes ($\text{Bq m}^{-2} \text{ d}^{-1}$) is based on the time markers, assuming constant sedimentation between events (Δt), on dry bulk density (ρ [g cm^{-3}]), and the individual sample thickness (d [m]):

$$F = \frac{A\rho d}{\Delta t} \quad (1a)$$

The variation in the flux of ^{60}Co with time in each sediment core is a function of both radionuclide discharge and scavenging factors. For each of our seven cores an average

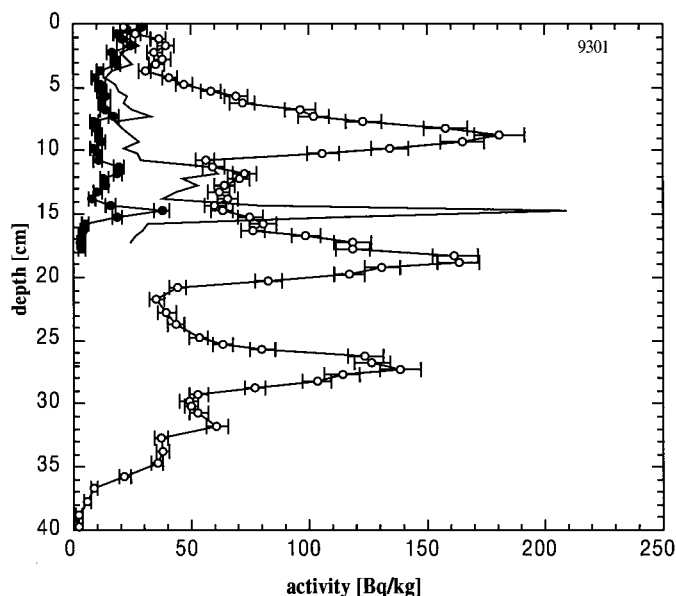


Fig. 3. Radionuclide depth distribution in core 9301 illustrating sediment dating on the basis of ^{137}Cs maxima of known age (\circ = decay corrected to the day of sampling with 2σ measuring error). ^{60}Co (\bullet = decay corrected to the day of coring, with 2σ errors) can be decay corrected to the time of deposition using the ^{137}Cs age scale and assuming constant sedimentation between time markers (continuous line).

flux for the period 1986 to the date of coring has been calculated and is shown in Table 2, with additional information on sedimentation rates, median grain size, and particle mineralogy. In spite of the significant differences in rate and type of sedimentation, the variance is small.

The maximum and the second lowest depositional rates (0.97 and $0.38 \text{ Bq m}^{-2} \text{ d}^{-1}$, respectively) were taken within $<500 \text{ m}$ of each other in the NE basin of the lake. This difference is due either to the relation between the time of sampling and effluent discharges of the nuclear reactor or to differences in local sediment deposition. The core (9401) with the highest average flux was taken immediately after a maximum discharge that occurred during the winter 1993/1994, whereas the core sampled in close vicinity was taken 2 yr later. Possibly the high ^{60}Co found in core 9401 can be explained by the lack of chemical mobilization associated with Mn cycling (Balistrieri et al. 1992, 1994). Anoxia occurs only during late summer and fall months and only in the NE basin. In core 9401, remobilization of ^{60}Co deposited during the winter/spring period could not occur because the core was taken at the end of May, before anoxia had occurred. ^{60}Co deposited in core 9601 had probably undergone some remobilization during anoxia.

In spite of these local variations, the variance is rather small (mean \pm SD = $0.52 \pm 0.23 \text{ Bq m}^{-2} \text{ d}^{-1}$). Disregarding the high value of $0.97 \text{ Bq m}^{-2} \text{ d}^{-1}$ results in a slightly lower mean and standard deviation ($0.45 \pm 0.12 \text{ Bq m}^{-2} \text{ d}^{-1}$). Multiplying the average flux with the surface area of sedimentation in Lake Biel then yields the total average deposition of ^{60}Co . The surface area of sedimentation can be assessed by subtracting from the total lake surface area the shallow

Table 2. Flux of ^{60}Co and information on particles.

Core location*	^{60}Co flux ($\text{Bq m}^{-2} \text{ d}^{-1}$)	Sedimentation rate (cm yr^{-1})	Median particle size (μm)	Quartz (%)	Feldspar (%)
9701	0.29	3.7	23.6	18	14
9301	0.44	1.4	8.8	21	7
9401	0.97	2.0	7.5	12	2
9601	0.38	1.7	8.9		
9602	0.48	2.0	14.5		
9404	0.65	1.5	11.6	9	1
9407	0.43	1.4	8.5	14	2
Trap	0.41		14.9	8	1

* See Fig. 1.

water portion where erosion negates sedimentation (Weiss 1979; Wright et al. 1980; Fig. 1, Table 1). The total averaged daily sediment flux in Lake Biel is estimated to be $1.57 \times 10^7 \text{ Bq d}^{-1}$. Comparison with the average daily discharges of the nuclear reactor ($3.03 \times 10^7 \text{ Bq d}^{-1}$ of ^{60}Co for the period 1986 to October 1996) yields an average deposition of 52%. Taking into account the analytic error ($\sim 10\%$), a 5% error on both density and sample depth determination, and a 10% error for time calculation, the relative error for the flux determination is 16% for ^{60}Co . The extent to which the seven cores represent the entire deposition surface can be estimated using the standard deviation of the mean, which is 44%. Rejection of the high value of $0.97 \text{ Bq m}^{-2} \text{ d}^{-1}$ reduces the error to 27%. Depending on the rejection or retention of the core with the highest average flux, ^{60}Co deposition in Lake Biel lies in the range of 30–70% (core included) or 30–55% (core rejected).

Seasonal variation—In cores with high temporal resolution, the variation of the ^{60}Co flux can be compared with monthly discharges from the nuclear reactor. An illustration of this comparison is given in Fig. 4, where the depth distribution of the ^{60}Co flux is displayed in terms of calendar dates. For direct comparison of the sediment deposition flux with the discharge from the reactor, monthly discharge activities (Bq month^{-1}) have been distributed evenly over the sedimentation area of the lake. In core 9301 (Fig. 4a), where the sampling interval was held constant at $0.5 \text{ cm sample}^{-1}$ and where the sedimentation rate fortuitously turned out to be 1.4 cm yr^{-1} , a seasonality in ^{60}Co scavenging becomes evident. During the warm period (particularly May–September), the flux into the sediment represents only 24–50% of the flux into the lake. During the winter period, the situation changes and sediment output fluxes increase to reach 60–170%. A second core (9602, Fig. 4b) shows the same seasonal dependence for ^{60}Co scavenging, although more pronounced for 1995 than for 1996. During the warm period, 30–75% of the discharge was transferred to the sediment, whereas during the cold period the proportion rose to 103–131%.

Particle concentration as a function of depth and season—The variation of particle concentration in the Aare River and Lake Biel water samples is quite significant (Fig. 5), ranging from 0.5 to 160 g m^{-3} in the river (with most measurements between 1 and 5 g m^{-3}) and from 0.2 to 5.4 g m^{-3} in the lake.

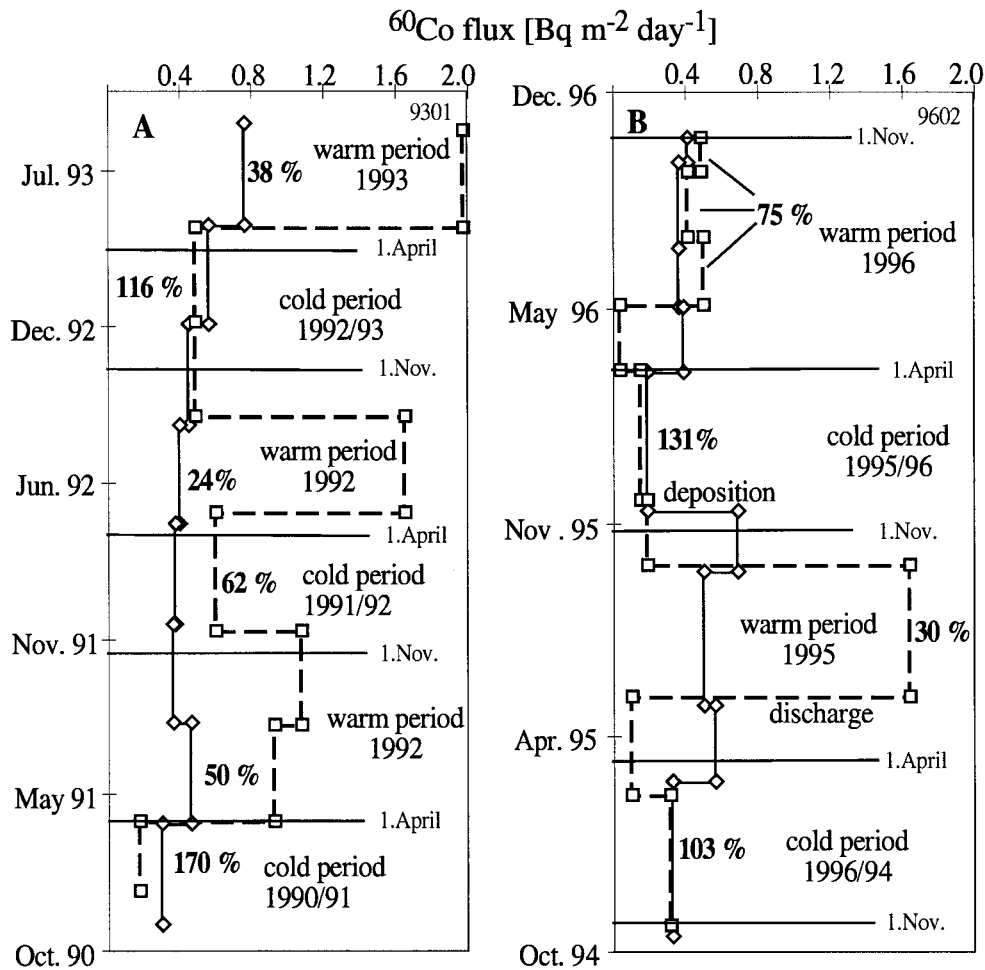


Fig. 4. Comparison of ^{60}Co fluxes into the sediment (continuous line with diamonds) with fluxes from the nuclear reactor into the lake (evenly distributed throughout the entire lake sedimentation area, dashed line with squares) as a function of depth/time in two cores. For typical periods, the proportion of ^{60}Co scavenged and transported into the sediments is indicated. As a general rule, scavenging is more efficient during the cold than during the warm period. All percentage values indicate the proportion of discharged ^{60}Co transferred to sediments.

Of importance for our interpretation is the concentration range in the lake as a function of depth and season. During both the cold and the warm period, particle concentrations tend to be higher in the uppermost and lowest 20-m sections of the lake. In general, particle concentrations reach their maxima during summer. A ^{60}Co scavenging model, which is based solely on particle concentration, would therefore predict higher scavenging efficiency during summer than during winter, the opposite to what we found in the sediment.

Intrusion model—One of the crucial factors for the fate of the radionuclides in Lake Biel is the depth at which the Aare River intrudes into the lake. Both the average residence time of the radionuclides and the biochemical environment of the radionuclides are mainly determined by the intrusion process.

Buoyancy is the driving force for many oceanic and limnologic processes, such as deep-water formation, gravity, or turbidity currents. A set of simple conservation equations can be used to describe the main dynamics of such phenomena.

These stream tube or plume models were first formulated by Smith (1975) and Killworth (1977), and they have been frequently applied to such simulations. A modified version of the model, proposed by Alendal et al. (1994) (Fig. 6) has been applied here. The conservation of mass, internal energy (heat), salt, and momentum along the stream direction, averaged over the cross-sectional area $A = wh$, where w and h are, respectively, the width and the height of the current, is given by

$$\frac{\partial(\rho AU)}{\partial y} = \rho_e E w U \quad (1b)$$

$$\frac{\partial(T\rho AU)}{\partial y} = \rho_e T_e E w U \quad (2)$$

$$\frac{\partial(S\rho AU)}{\partial y} = \rho_e S E w U \quad (3)$$

$$\frac{\partial(\rho AU^2)}{\partial y} = Ag(\rho - \rho_e)\sin \alpha - C_D \rho w U^2. \quad (4)$$

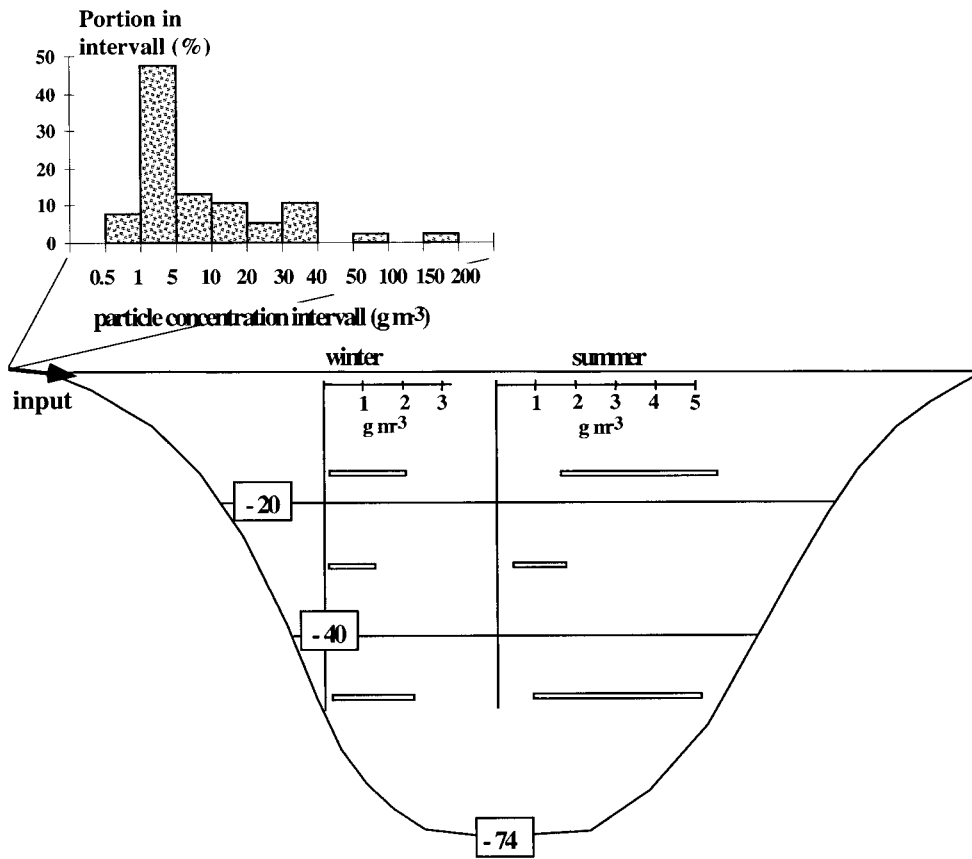


Fig. 5. Distribution of particle concentration (cutoff between 0.45 and 1.0 μm) in the inflowing Aare water (upper inset) and within the lake (bars indicating the range of particle concentration as a function of season and depth). In spite of the large variance, particle concentrations at any depth are higher during the summer than during the winter.

Here ρ , S , and T represent the density, salinity, and temperature of the current, respectively. U denotes the mean current velocity, E is the entrainment factor, C_D is the drag coefficient, and α is the angle of the lake bottom at the intrusion. Variables with subscript e are related to the ambient lake water.

The Coriolis force was neglected in these equations be-

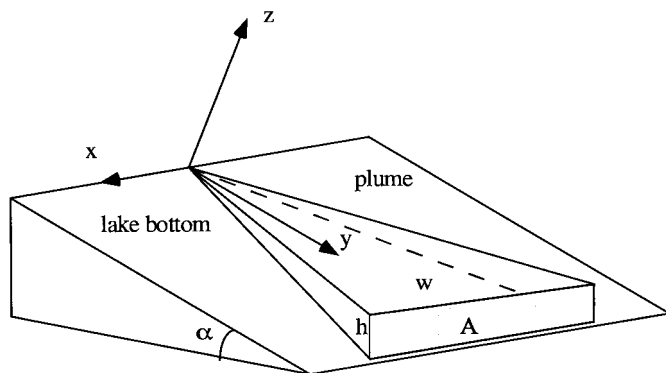


Fig. 6. Physical configuration and coordinate system of the plume model used for simulating the intrusion of Aare water into Lake Biel. See text for symbols.

cause its influence on the intrusion depth can be assumed to be marginal in this case. The equation for the state of lake water from Chen and Millero (1986) was used to compute the density ρ of lake and river water as a function of salinity and temperature. To close the system of ordinary differential equations, the similarity assumption was made, i.e., $w/h = \text{constant}$ (Alendal et al. 1994). In our study, this parameter is determined by the geometry of the Aare River at the inflow, i.e., ≈ 100 . The entrainment parameter E was estimated from field measurements in Lake Urnen; $E = 0.015\text{--}0.025$, a result similar to the value of 0.02 derived from laboratory experiments (Christodoulou 1986). The average friction parameter C_D was calculated from the parameterization of Alendal et al. (1994):

$$C_D = \left(\frac{RiE}{2} \right)^{2/3}, \quad (5)$$

which yields $C_D \approx 0.06$ (where Ri is the Richardson number). The slope angle α was determined from the lake morphology ($\alpha = 0.035$).

The variables for the lake water (i.e., T_e and S_e) were taken from experimental CTD measurements at the deepest point of the lake, far away from the river inflow, to guarantee genuine ambient values without distortion of river water. The

Intrusion Depth [m]

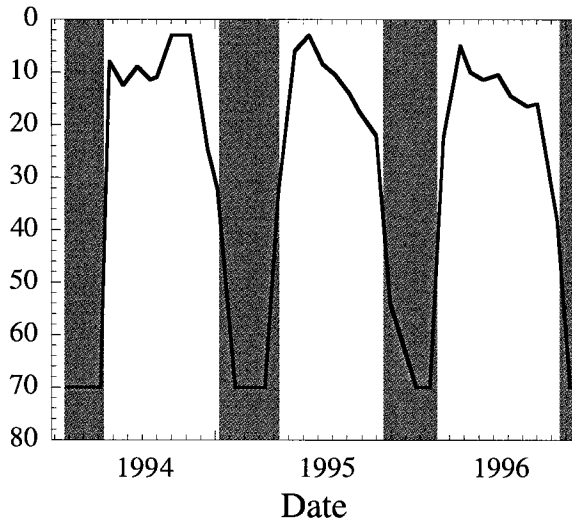


Fig. 7. Simulated intrusion depths of the Aare water into Lake Biel for January 1994–December 1996. With the development of a stratification, the intrusion depth varies between 3 and 20 m. During the annual overturn in winter (gray), the Aare intrudes into the deep hypolimnion.

initial values of U , T , S , and ρ were provided by daily measurements at the inflow in the lake. The intrusion depth computed by the model is defined as the depth at which the river plume and the ambient lake water have the same density (i.e., $\rho = \rho_c$).

Validation of the model for the Lake Biel case study is based on tracer experiments. The general behavior and the seasonal development of the intrusion depth can be seen from simulations for 1994–1996 (Fig. 7). A clear seasonal development of the intrusion depth can be observed; during winter (i.e., December–March) the Aare River intrudes either into the deepest point of the lake or below the seasonal thermocline. This phenomenon is driven mainly by the high salinity of the Aare; temperatures during this period are close to the temperature of maximum density. During the rest of the year (i.e., April–November), the Aare River intrudes above the seasonal thermocline because temperature effects dominate the intrusion; the temperature in the thermocline in summer is always much lower than that of the Aare River. This 2 fold physical behavior of Aare water in the lake has consequences for radionuclide transport. During summer (warm period), Aare water intrudes into the upper lake section and has the ability to cross the lake, mixing with only a fraction of the total lake volume. The estimated cross-flow for Aare water in the lake was 0.14 km^3 , compared to the total volume of 1.2 km^3 . The travel time in the lake (as a function of Aare runoff) would be 5–7 days. During transit the activity per volume decreases due to mixing with lake water, due to dispersion and due to the removal of radionuclide carrying particles. Even though particle concentrations are at a maximum during summer, in view of slow adsorption kinetics (Li et al. 1984) and the high proportion of dissolved radionuclides entering the lake, scavenging remains inefficient during this period. Lakewide deposition of

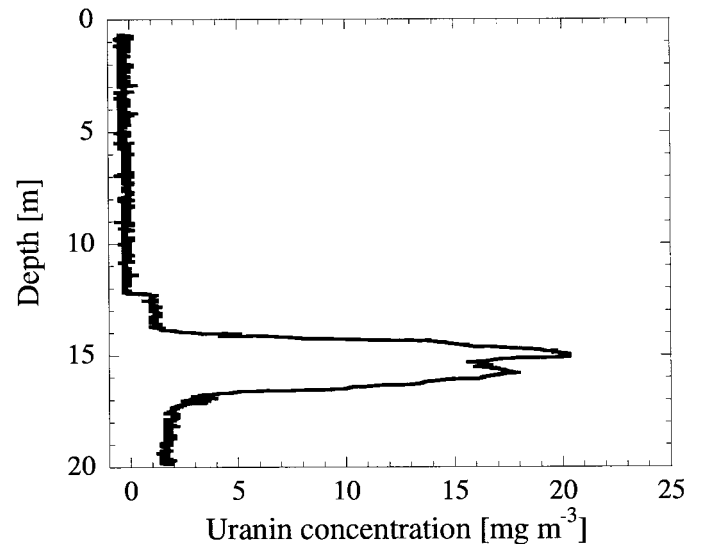


Fig. 8. Uranin concentration profile measured in Lake Biel on 20 August 1996, 1 d after uranium addition to the Aare at the Mühleberg reactor (cf. Fig. 1). The measured intrusion depth ($\sim 15 \text{ m}$) corresponds well with modeled results.

radionuclides throughout the summer indicates that a portion of the radionuclide input is distributed throughout the lake, probably as a result of inflow-induced current systems with counterclockwise water circulation operating at all depths (Nydegger 1976b; Wright and Nydegger 1980).

During winter the river water intrudes into deeper sections of the lake. As Aare runoff drops to an average of $\sim 115 \text{ m}^3 \text{ s}^{-1}$ during the winter period, residence times can rise up to 120 d, which allows more time for absorption processes and thus explains more efficient scavenging in spite of lower particle concentrations.

Experiments to test the model—Uranin: During a regular discharge of the nuclear power plant Mühleberg, 10 kg of uranin (sodium fluorescein) was introduced into the Aare River at the reactor site on 19 August 1996 at 2240 h. The measured breakthrough of uranin at Hagneck occurred between 0400 and 0900 and coincided well with the modeled breakthrough. During this period, samples were taken to measure both stable and radioactive Co and particle concentration. The uranin distribution in the lake as a function of location and depth was the subject of a detailed investigation using a fluorometer. A maximum was found at a depth of 14–16 m (Fig. 8). By the early afternoon of 20 August 1996, the uranin-carrying Aare water had covered an area of approximately 7.1 km^2 , or 18% of the lake surface (Fig. 9). The distribution was homogeneous around the two inlets formed by the Aare River. The right-lateral drift measured by others (Nydegger 1976b; Wright and Nydegger 1980) can be seen in the dislocation of the uranin maximum toward the NE. On 21, 22, and 27 August we were able to locate the maximum of the uranin as a function of position and depth along a transect between the inlet and the outlet (Fig. 1). These results confirmed earlier measurements, where a ^{60}Co maximum, related to a known release from the nuclear

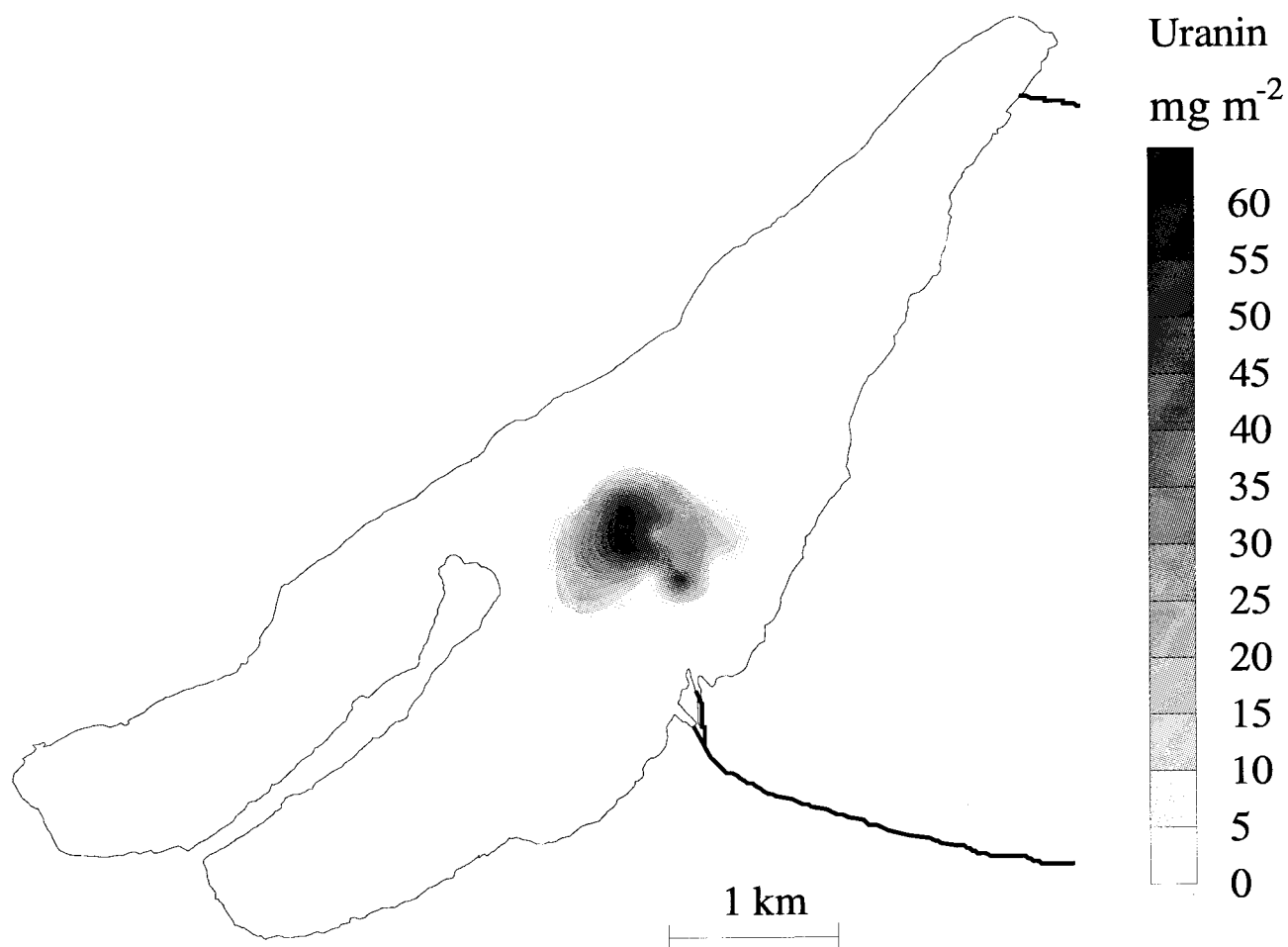


Fig. 9. Contour plot of uranium fluorescence in Lake Biel on 20 August 1996. The individual uranium profiles were integrated vertically and interpolated horizontally to obtain isoconcentration areas. Contrary to previous expectations, the tracer cloud did not follow the northeastern border of the lake but spread evenly into the central part of the lake.

reactor, was found at the outlet of Lake Biel 7 days after discharge.

Results obtained during the dye tracer experiment allow validation of the mathematical intrusion model. The model reliably predicted the intrusion depth on 20 August (~ 15 m, Fig. 8).

Sediment traps: Results obtained for the period, July 1996–March 1997, further support the model. Between July and October, during the warm period, the average particle flux of the upper, intermediate, and lower sediment traps varied only between 12 and 17.3 $\text{g m}^{-2} \text{d}^{-1}$ (Fig. 10A); the variance in the ^{60}Co flux (0.37 – 0.45 $\text{Bq m}^{-2} \text{d}^{-1}$) was likewise small. The slightly higher particle and ^{60}Co flux of the upper trap relative to the intermediate trap can be explained by the decomposition of a portion of the particulate organic matter during descent.

Similarly, the slight increase in both particulate and ^{60}Co flux in the lower as compared with the intermediate trap could have its origin in bottom sediment resuspension (Bloesch 1995). To evaluate the importance of resuspended material, the flux of the natural radionuclide ^7Be with a half-

life of only 53 days is also given in Fig. 10. During the summer, the flux of ^7Be decreases as a function of depth. This decrease reflects decay during particle descent but also the incorporation of resuspended surface sediment with reduced ^7Be activity.

Because both particle and ^{60}Co flux in Lake Biel are dominated by the Aare River, the similarity in fluxes found in the three traps indicates that the Aare River water remains above the upper trap, thus within the epilimnion. The average ^{60}Co flux of the three sediment traps for the period July–September (0.41 $\text{Bq m}^{-2} \text{d}^{-1}$), if assumed representative of the entire lake sediment surface, yields a total deposition rate for the summer of 1.2×10^7 Bq d^{-1} . Comparison with the average release of the nuclear reactor (1.9×10^7 Bq d^{-1}) allows a second estimation of the ^{60}Co deposition, which is independent of the one relying on sediment cores. The slightly higher result of 64%, compared with 52%, on the basis of the sediments is not surprising. The shorter inventory period and the extrapolation from a single location to the entire lake sediment surface could have produced either a negative or a positive error, but overestimation of the sed-

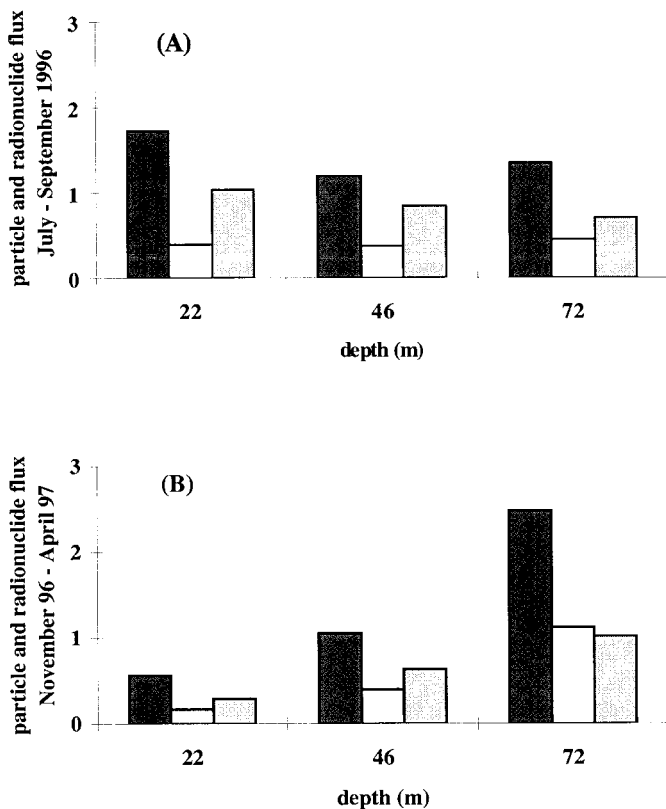


Fig. 10. Seasonal average particle flux (dark gray, g m⁻² d⁻¹, reduced by a factor of 10) and seasonal average flux of artificial ⁶⁰Co (white, Bq d⁻¹ m⁻²) and natural ⁷Be (light gray, Bq d⁻¹ m⁻², reduced by a factor of 10) in three sediment traps positioned below the thermocline, within the hypolimnion, and 2 m above ground during the warm (A) and cold (B) periods.

imentation rates on the basis of trap results (Bloesch 1995) tends to generate higher fluxes for trap-based inventories.

The situation changes quite distinctly during the cold period. Both average particle and radionuclide fluxes (⁶⁰Co and ⁷Be) increase with depth (Fig. 10B). For the period November 1996–March 1997, the particle flux increased from 5.6 to 24.8 g m⁻² d⁻¹; likewise, the ⁶⁰Co and ⁷Be fluxes increased from 0.17 to 1.1 and from 2.9 to 10.2 Bq m⁻² d⁻¹, respectively. Such increases cannot be explained by higher sediment resuspension because resuspended sediment would have a negligible amount of ⁷Be (because of its short half-life). The sediment trap data, therefore, give strong support to our intrusion model, which predicts that Aare water, including its particle and radionuclide load, sinks deeper into

the lake and delivers significantly more material to the lower sediment trap.

⁶⁰Co concentration in lake water—The intrusion model can predict the activity of those radionuclides with known input exclusively from the Aare River. This prediction is possible for ⁶⁰Co, but the low anticipated activities and the need to treat several hundred liters of water do not permit a large number of samples. Direct comparison of a single activity measurement with the known input from the reactor is only possible at the end of the winter period. During the winter, the lake is replenished with Aare water, which sinks into the deeper part of the lake. The period required to replace the entire lake volume is based on the known runoff of all tributaries and can be set equal to the related discharge period of the nuclear reactor (Table 3). Three high-volume samples were taken to test this hypothesis, two on 28 March 1996 at 6 and 50 m and one on 2 February 1997. In 1996, the activity increased with depth from 0.33 to 0.54 Bq m⁻³; in 1997, the activity was 0.3 Bq m⁻³. The modeled activity based on discharges by the nuclear reactor is 0.5 Bq m⁻³ for the earlier samples and 1.1 Bq m⁻³ for the later samples. Removing 50% of the calculated activity in the water column by sedimentation yields modeled activities of 0.25 and 0.55 Bq m⁻³. In view of the complexity of the system and the number of assumptions made, the agreement of measured and estimated activities can be considered satisfactory.

Summary

Lake Biel, a Swiss lowland lake dominated by the River Aare, is an ideal site to study the combined effects of river water intrusion into the lake and geochemistry on radionuclide transfer and deposition because of the presence of a unique radio tracer with known input and a low water residence time.

The residence time of the radionuclide-carrying Aare water depends essentially on the physical state of Lake Biel. During summer, a stable stratification develops, and Aare water remains in the epilimnion and crosses the lake within 6–7 d. During winter, the Aare water intrudes into deeper sections of the lake and essentially mixes with the entire lake volume. Depending on river runoff, residence time can reach up to 4 months.

The behavior of Aare water in the lake can be modelled on the basis of conservation of mass, internal energy (heat), salt, and momentum. Validation of the model was possible using results from the tracer experiment.

Table 3. Comparison between calculated and measured activities of ⁶⁰Co in Lake Biel for samples collected at the end of the winter period.

Date	Activity ⁶⁰ Co (Bq m ⁻³)	Error 2σ (%)	Sampling depth (m)	Lake volume (km ³)	Fill period	Discharge (Bq)	Calculated ⁶⁰ Co (Bq m ⁻³)	
							Water column	50% in sediments
28 Mar 96	0.3	30	6	1.18	Dec 95–Mar 96	5.4 × 10 ⁸	0.5	0.25
28 Mar 96	0.5	40	50	1.18	Dec 95–Mar 96	5.4 × 10 ⁸	0.5	0.25
6 Feb 97	0.3	14	45	1.18	Dec 96–Jan 97	9.5 × 10 ⁸	1.1	0.6

The physical model explains the major differences in radionuclide scavenging between the cold and the warm period. During the cold period, the river water and the associated radionuclide and particle load remain longer in the lake, giving more time for radionuclide scavenging, than during the warm period, when residence times drop below 1 week. Lower scavenging occurs in spite of higher particle concentrations during the warmer period. The flux of particles and radionuclides into sediment traps positioned at different depths is similar during the warm period. During the cold period, when Aare water carries particles and radionuclides to greater depth, the deeper sediment traps recorded higher particle and radionuclide fluxes.

On the basis of sediment cores, the proportion of radiocobalt scavenged in the lake has been estimated to range between 30% and 55%. Sediment traps have confirmed this result for summer 1996.

References

- ALBRECHT, A., AND OTHERS. 1997. The transport of Cobalt in Lake Biel—in the footsteps of the Aare River. *EAWAG News* **43**: 21–24.
- , P. REICHERT, J. BEER, AND A. LÜCK. 1995. Evaluation of the importance of reservoir sediments as sinks for reactor-derived radionuclides in riverine systems. *J. Environ. Radioact.* **28**: 239–269.
- , R. REISER, A. LÜCK, J. M. STOLL, AND W. GIGER. 1998. Radiocesium dating of sediments from lakes and reservoirs of different hydrological regimes. *Environ. Sci. Technol.* **32**: 1882–1887.
- ALENDAL, G., H. DRANGE, AND P. HAUGAN. 1994. Modelling of deep-sea gravity currents using an integrated plume model. *Geophys. Monogr.* **85**: 237–246.
- BALISTRERI, L. S., J. W. MURRAY, AND B. PAUL. 1992. The biogeochemical cycling of trace metals in the water column of Lake Sammamish, Washington: Response to seasonally anoxic conditions. *Limnol. Oceanogr.* **37**: 529–548.
- , ———, AND ———. 1994. The geochemical cycling of trace elements in a biogenic meromictic lake. *Geochim. Cosmochim. Acta* **58**: 3993–4008.
- BLOESCH, J. 1995. Mechanisms, measurement and importance of sediment resuspension in lakes. *Mar. Freshwater Res.* **46**: 295–304.
- BUFFLE, J. [ED.]. 1988. Complexation reactions in aquatic systems: An analytical approach. Ellis-Horwood.
- , AND H. P. VAN LEEUWEN [EDS.]. 1992. Environmental particles. Lewis.
- CHEN, C. T., AND F. J. MILLERO. 1986. Precise thermodynamic properties for natural waters covering only the limnological range. *Limnol. Oceanogr.* **31**: 657–662.
- CHRISTODOULOU, G. C. 1986. Interfacial mixing in stratified flows. *J. Hydrol. Res.* **24**: 77–92.
- GARNIER, J.-M., M. K. PHAM, P. CIFFROY, AND J.-M. MARTIN. 1997. Kinetics of trace metal complexation with suspended matter and filtrable ligands in fresh water. *Environ. Sci. Technol.* **31**: 1597–1606.
- KILLWORTH, P. D. 1977. Mixing on the Wedell sea continental slope. *Deep-Sea Res.* **24**: 427–448.
- LI, Y.-H., L. BURKHARDT, M. BUCHHOLTZ, P. O'HARA, AND P. H. SANTSCHI. 1984. Partition of radiotracers between suspended particles and seawater. *Geochim. Cosmochim. Acta* **48**: 2011–2019.
- NYDEGGER, P. 1976a. Strömungen in Seen. Untersuchungen in situ und an nachgebildeten Modellseen. *Beitr. Geol. Schweiz Kl. Mitt.* **66**: 142–177.
- . 1976b. Untersuchungen über Feinstofftransport in Flüssen und Seen, über Entstehung von Trübungshorizonten und zuluftbedingten Strömungen im Breinzersee und einigen Vergleichsseen. *Beitr. Geol. Schweiz Hydrol.* **16**: 68–89.
- O'MELIA, C. R., AND K. S. BOWMAN. 1984. Origins and effects of coagulation in lakes. *Schweiz. Z. Hydrol.* **46**: 65–85.
- REICHERT, P. 1994. AQUASIM—a tool for simulation and data analysis of aquatic systems. *Water Sci. Technol.* **30**: 21–30.
- REISER, R., H. TOLJANDER, A. ALBRECHT, AND W. GIGER. 1997. Alkylbenzenesulfonates in recent lake sediments as molecular markers for the environmental behavior of detergent-derived chemicals, p. 196–212. *In* R. P. Eganhouse [ed.], *Molecular markers in environmental geochemistry*. American Chemical Society.
- SMART, P. L. 1982. A review of the toxicity of 12 fluorescent dyes used for water tracing. *Beitr. Geol. Schweiz Hydrol.* **28**: 101–112.
- SMITH, P. C. 1975. A streamtube model for bottom boundary currents in the ocean. *Deep-Sea Res.* **22**: 853–873.
- SPARKS, D. L. 1995. Kinetics of metal sorption reactions, p. 35–58. *In* H. E. Allen, C. P. Huang, G. W. Bailey, and A. R. Bowers [eds.], *Metal speciation and contamination of soil*. Lewis.
- STUMM, W., AND J. J. MORGAN. 1996. *Aquatic chemistry; chemical equilibria and rates in natural waters*, 3rd ed. Wiley.
- TSCHUMI, P. 1977. Eutrophierung, Primärproduktion und Sauerstoffverhältnisse im Bieler See. *Gas Wass. Abwass.* **57**: 245–252.
- WEILENMANN, U., C. R. O'MELIA, AND W. STUMM. 1989. Particle transport in lakes: Models and measurements. *Limnol. Oceanogr.* **34**: 1–18.
- WEISS, H. P. 1979. Die Oberflächensedimente des Bieler Sees. *Eclogae Geol. Helv.* **72**: 407–424.
- WRIGHT, R. F., A. MATTER, M.-SEHWEINGRUBER, AND U. SIEGENTHALER. 1980. Sedimentation in Lake Biel, an eutrophic lake in northwestern Switzerland. *Schweiz. Z. Hydrol.* **42**: 101–126.
- , AND P. NYDEGGER. 1980. Sedimentation of detrital matter in lakes: Influence of currents produced by inflowing rivers. *Water Resour. Res.* **16**: 597–601.

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