

The effect of Ozone on Waters Containing Chlorine or Bromide

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Many drinking water treatment plants, especially in Switzerland and neighboring countries, use ozone either for disinfection or to improve the chemical quality of the water.

Chlorine is also often used in a preceding or subsequent treatment step. However, ozone and chlorine react with one another, resulting in a decrease in efficacy of both oxidants. How fast does this reaction occur under practical water treatment conditions?

Outside of Switzerland waters sometimes contain traces of bromide, which also reacts with ozone during drinking water treatment. The advantages and disadvantages of using ozone in seawater (which contains large amounts of bromide) for shellfish depuration or cooling water antifouling are under discussion. Are the bromide ions not simply oxidized to «active» bromine? Such questions are of interest since «active» bromine not only interferes in the measurement of ozone concentrations, but can also produce undesirable brominated compounds, such as bromoform in the presence of natural organic compounds.

In this article we will briefly describe how basic, experimentally accessible, kinetic data can be used to predict the behavior of chemical oxidants used in water treatment.

1. Interaction of Ozone with Chlorine

When gaseous chlorine is added to water it rapidly hydrolyzes; therefore, «active» chlorine exists mainly as hypochlorous acid (HOCl) (see Fig. 1). HOCl is a weak acid which is partly dissociated to the hypochlorite ion (OCl⁻) at the pH of drinking water. Our kinetic measurements show that ozone reacts only with OCl⁻ and not with HOCl. Thus, the rate with which ozone reacts with aqueous chlorine increases proportionately to the degree of dissociation, α , of the hypochlorous acid; that is, it increases by a factor of 10 per pH unit up to pH 7:

$$-\frac{d[O_3]}{dt} = 160 \text{ M}^{-1}\text{s}^{-1} \alpha [\text{HOCl}]_{\text{tot}} [O_3]$$

where $\alpha = \frac{1}{1+10^{(7.5-\text{pH})}}$

As shown in Fig. 1, ozone reacts with hypochlorite ion by two different pathways. Only about 23% of the OCl⁻ ends up as undesirable chlorate (ClO₃⁻); the main pathway (77%) leads to relatively non-toxic chloride.

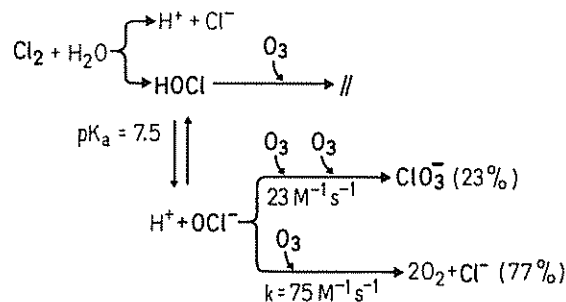


Fig. 1: Reactions of ozone with aqueous chlorine.

Using the kinetic rate law given above, we calculated the half-lives of ozone or chlorine in the presence of one another (see Fig. 2). For example, if the ozone concentration is kept at 1 mg/l, which lies in the range normally used in water treatment plants, the half-life of the chlorine at pH 7-8 will be about 10 minutes. The same is true for the half-life of ozone if the chlorine concentration remains constant at about 1.5 mg/l Cl_{2,tot}. The reaction is slow when compared with disinfection processes and, therefore, should not interfere significantly with the latter. On the other hand, the residual chlorine concentration in the water distribution network will be reduced, increasing the risk of a re-infection.

For specialists in this field, we should note that combined chlorine, formed by chlorination of ammonium-containing waters, reacts somewhat more slowly with ozone than does hypochlorite ion [1].

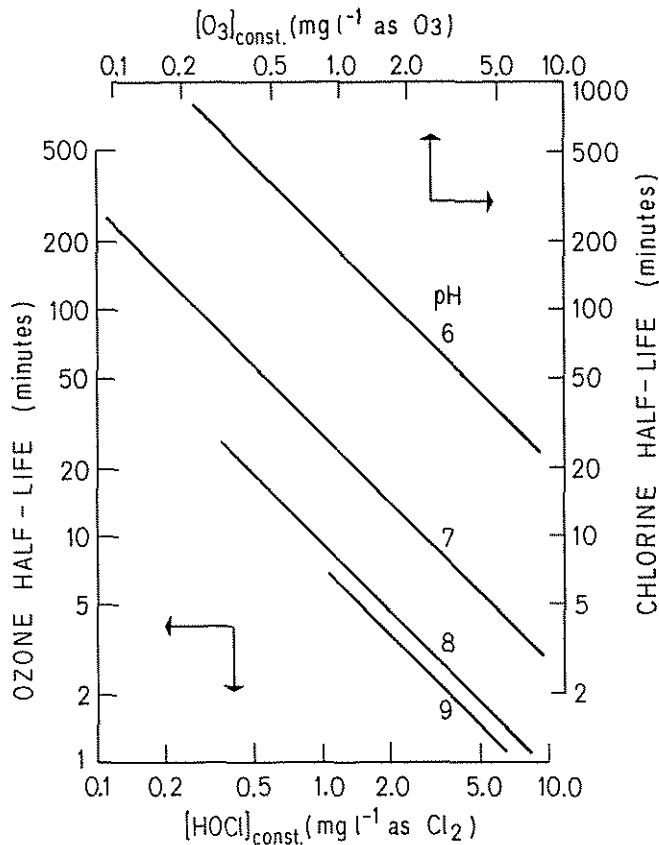


Fig. 2: Stability of active chlorine and ozone in water in the presence of each other. Lower & left axes: half-life of ozone assuming the concentration of chlorine remains constant. Upper & right axes: half-life of chlorine assuming the concentration of ozone remains constant.

2. Interaction of Ozone with Bromide Ion

Many waters which are ozonated for disinfection, such as drinking water, swimming pool water, and power plant cooling water, contain at least traces of bromide (see Table 1). Ozone oxidizes bromide to «active» bromine (HOBr/OBr⁻), which acts as a secondary oxidant and disinfectant. This active bromine is then further oxidized to bromate, the stable end product. Using model reaction mixtures for the kinetic measurements, we were able to establish and quantify the reaction scheme shown in Figures 3 and 4 [2].

Fig. 3: Reactions of ozone with aqueous bromine and bromide ion.

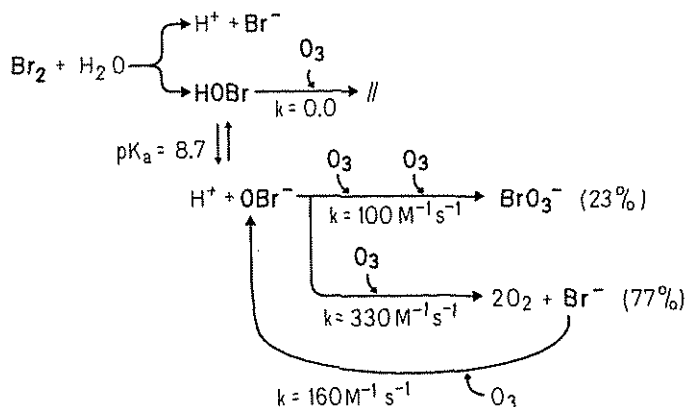


Table 1: Typical bromide concentration in various waters [2]

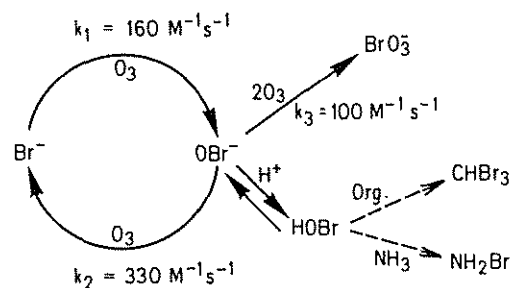
	mg/l Br ⁻
Sea Water	65
Lake of Zurich	0.006
River Rhine at Basle	0.03
River Rhine at Karlsruhe	0.2
River Rhine at Netherlands	0.06–0.8
Five major rivers in USA	0.004–0.08
Ground water (Belgium)	0.03–0.3
Rivers (Belgium)	0.02–0.2
Various Mineral Waters (Switzerland, France, Fed. Rep. Germany, Belgium)	≤ 2.4

Analogous to the reaction with active chlorine, only the hypobromite ion reacts with ozone, and only a minor part (23%) of this reaction leads directly to bromate ion (BrO₃⁻). The main pathway again leads to halide ion, via peroxide formation and O₂ loss. However, in contrast to chloride, bromide can be oxidized back to hypobromite by additional ozone. The result is a small chain reaction, whereby ozone is catalytically destroyed and the formation of bromate strongly inhibited (see Fig. 4).

Of primary interest is the concentration of active bromine which builds up during the ozonation of a bromide-containing water. A simple kinetic computer model and subsequent experimental verification gave the concentration-time profiles shown in Fig. 5. After only several minutes of continuous ozonation at 1 mg/l, a large portion of the initial bromide was oxidized to active bromine. However, since this exists in a dynamic equilibrium with bromide, only a certain percentage of the total bromine can be present as active bromine. This percentage is particularly low at high pH values where the regeneration of bromide is not inhibited by the protonation of the OBr⁻ to HOBr. Nevertheless, in the pH range typical of drinking and cooling waters (pH 7–8), a substantial portion of the initial bromide exists as active bromine over a rather long ozonation period.

The presence of active bromine in ozonated waters causes certain problems. Conventional analytical methods for the determination of residual ozone, which are usually based on oxidation, give a false positive response to active bromine. Moreover, aqueous bromine will brominate organic water constituents. Fig. 6 gives direct experimental evidence for the formation of bromoform upon addition of aqueous ozone to mixtures of humic acid and bromide ion. However, the extent of formation of halogenated organics appears to be less with ozonation than with chlorination. The formation of brominated phenols can be excluded since data from rate constant catalogs we have established [3] show that phenols react over 1000 times faster than bromide; thus, they would

Fig. 4: Chain reaction of ozone in bromine- and bromide-containing water, including further reactions of hypobromous acid and hypobromite ion.



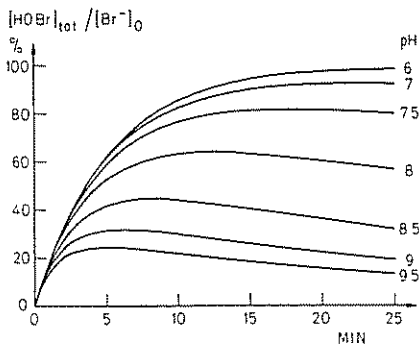


Fig. 5: Calculated active bromine concentrations during continuous ozonation of bromide-containing water at 1 mg/l ozone.

be oxidized before significant amounts of active bromine could be formed. This is the case even in seawater, despite the fact that the high bromide concentration (65 mg/l) limits the lifetime of the added ozone to only 6 seconds. In this respect, ozonation of seawater has an advantage over chlorination, even though active bromine is produced as a secondary oxidant in both cases.

These considerations demonstrate that kinetic data can be used to make predictions on the behavior of chemical oxidants and disinfectants used in water treatment, even when complex interactions must be considered. In a separate communication we will show how certain reaction pathways of atmospheric ozone can also be better understood and quantified by using corresponding kinetic data.

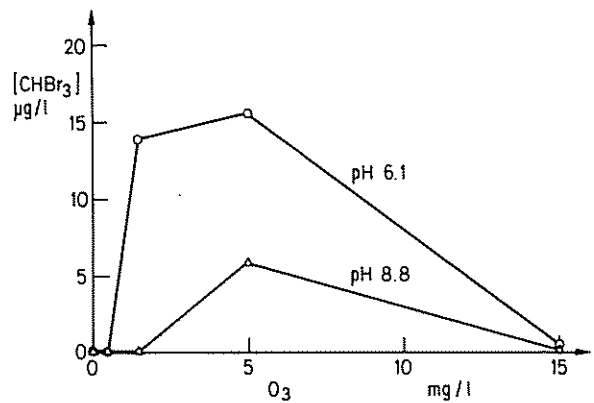


Fig. 6: Bromoform concentration measured 21 hours after addition of ozone to bromide- and humic acid-containing solution $[Br^-]_0 = 1$ mg/l; $[humic\ acid]_0 = 2$ mg/l; $[HCO_3^-] = 2$ mmol/l.

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Suspended Particles in Lakes

Michael Sturm

Introduction

Particles are excellent tracers to study geobiochemical processes in the natural environment. Knowledge of processes which lead to particle formation and descriptions of their composition and distribution in an aquatic system, therefore, increase our insights into those mechanisms which control that particular system.

Suspended material (= particles, suspensoids) in a lake may originate from different sources: the atmosphere, river input, land slides and formation processes within the lake itself (Fig. 1). Accordingly, suspended particles vary greatly in composition and represent a wide range of sizes: distinct single particles (rock fragments, individual crystals, algal cells, etc.) or aggregates (coagulates, fecal pellets, etc.). Due to differences in the catchment area, the tributaries, basin morphology, and trophic state, variable amounts of particles from an individual source will be contributed to a lake. In alpine and perialpine (alpine foreland) lakes, the sources of suspensoids are either allochthonous including all material carried to a lake by its tributaries, by avalanches and land slides, etc. or autochthonous, i. e. particle formation within the water column including particles formed by biological processes, precipitation, coagulation, etc. Material from both sources may be secondarily reworked in the lake by slumping and resuspension processes.

Suspended particles in a lake can be described quantitatively and/or qualitatively. Within the water column, the occurrence of suspensoids is quantified by their concentration ($mg \cdot l^{-1}$) and the flux to the lake bottom by their mass per area and time ($g \cdot m^{-2} \cdot d^{-1}$). Final deposition of particles on the

lake floor is expressed by the annual sedimentation rate ($cm \cdot y^{-1}$) or the annual rate of accumulation ($g \cdot m^{-2} \cdot y^{-1}$). Some of the different techniques which are applied to determine these parameters are schematically shown in Figure 2. The qualitative description of particles may include the characterization of particles by their shape and size, by their chemical composition or by their position within the biological system. Several techniques may be applied to measure particle grain sizes (Coulter Counter, Sedigraph), bulk particle chemistry (Atomic Adsorption Spectrometry, Neutron Activating Analysis, X-ray diffraction) and single particle composition (Scanning Electron microscopy/X-ray spectrometry).

1. Particle concentration

Comparisons of particle concentrations in a number of alpine and perialpine lakes show in general a mean annual, pelagic, epilimnetic suspended material concentration of 2–3 $mg \cdot l^{-1}$ (Table 1). Regardless of their trophic state, all lakes develop lower concentrations during the winter months and higher concentrations in the spring and summer. Average uniform values are exceeded in lakes with high allochthonous input (Lake Ammer: 21 $mg \cdot l^{-1}$ [4]) or near the mouth of main tributaries (Lake Brienz: 25 $mg \cdot l^{-1}$ [7]). Moreover, maximum concentrations may occur during periods of high productivity during the spring in the epilimnion of a lake (Lake Greifen: 7.4 $mg \cdot l^{-1}$ [9]) and by resuspension processes at the sediment/water interface. However, water profiling with transmissometer or nephelometer and short-period water sampling show that such peaks of high concentrations are rapidly reduced (within hours or a few days) to the seasonally varying mean background concentration of particles in a lake [7, 18].

Fig. 1:
Sources and sinks of suspended particulate matter in a lake (modified after [13]).

RIV = influx by rivers, slumps, avalanches etc.
 AIR = atmospheric influx by wind, rain etc.
 AUT = autochthonous material (bioproduction, chemical precipitation etc.)
 DIS = dissolution of particles
 RES = resuspended material (bottom-currents, subaquatic slumps etc.)
 UPW = influx by upwelling of material (groundwater, subaquatic fountains etc.)
 OUT = outflow of particles

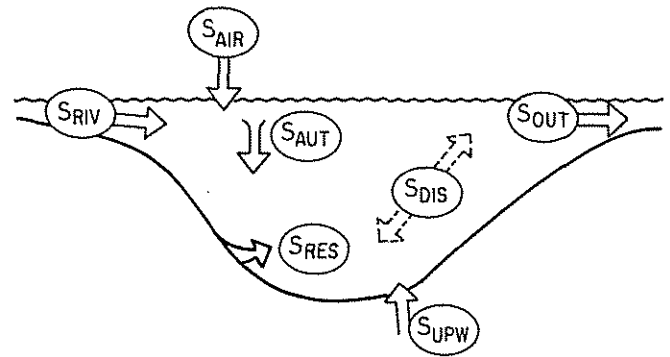


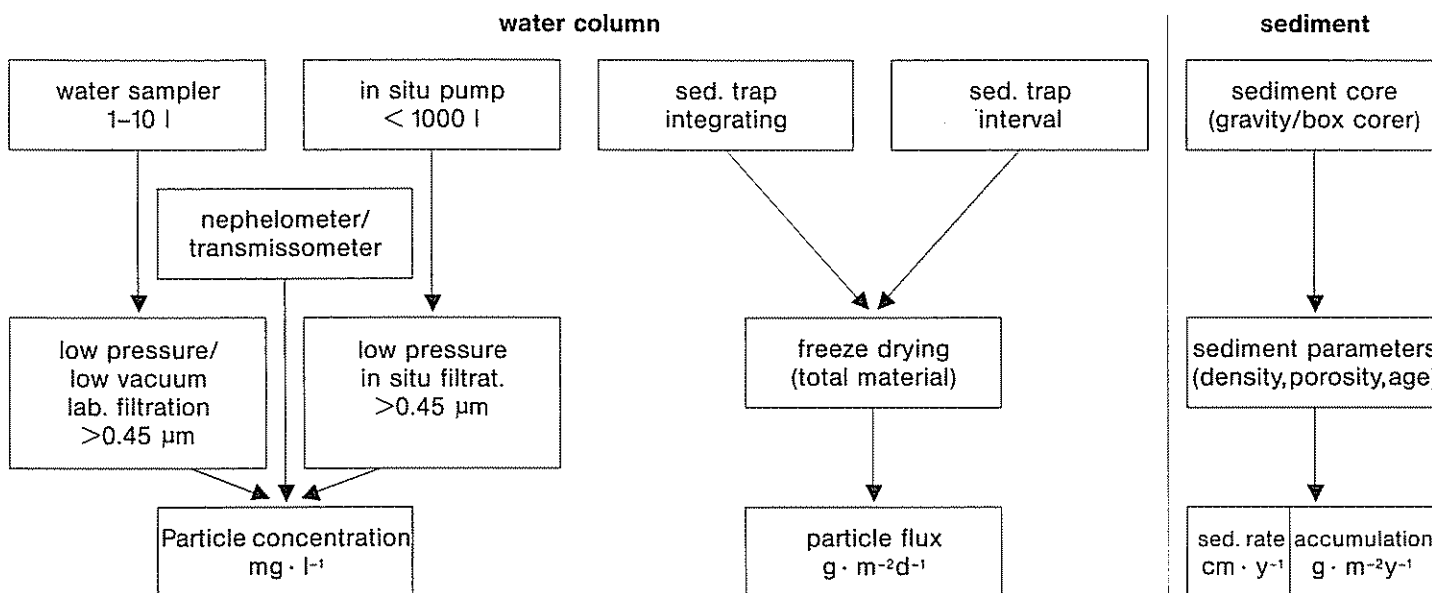
Table 1:

Particle and sediment data from alpine and peri-alpine lakes

Name of lake	Surface area (km ²)	Max. water Depth.(m)	Character	Part. concentration (mg · l ⁻¹)	Part. flux (g · m ⁻² d ⁻¹)	Sed. rate (cm · y ⁻¹)
L. Geneva [1]	582	310	oligo/mesotrophic large tributaries	1.8 (0.9–2.5)	3.3 (1.8–6.2) (Dec.–Aug.)	0.5 [14]
9 km of delta				–	9 (2.8–13.7) (Dec.–Aug.)	–
L. Constance [2]	475	252	oligo/mesotrophic large tributaries	3 (0.4–5.5) [11]	2 (0.2–19)	0.45 [3]
L. Zurich [3]	65	140	meso/eutrophic minor tributaries	2.2 (0.4–4)	2 (0.7–7)	0.35 [11]
L. Ammer/Germany [4]	47	82	meso/eutrophic large tributary	21 (17–23)	–	–
L. Biel [5]	39	75	eutrophic large tributary	2.5 (0.7–5.6)	5 (1–17)	1.1
L. Zug [6]	38	197	eutrophic minor tributaries	–	2 (0.2–9.3)	0.35
L. Brienz [7]	30	261	oligotrophic large tributaries	25 (4.3–54.7) (2 km off delta)	–	>2
L. Traun/Austria [8]	26	191	oligotrophic large tributaries	2.3 (1.2–2.9)	–	0.4 [3]
L. Greifen [9]	9	32	eutrophic minor tributaries	3.2 (1–7.4) (spring) [17]	6 (1.8–24)	0.4 [3]
Bay of Horw (L. Lucerne) [10]	2	72	mesotrophic minor tributaries	1.6 (0.3–4) [3]	3 (0.4–13.8)	0.4
L. Rot [10]	0.5	16	eutrophic no tributaries	–	3 (0.4–13.8)	0.37

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Fig. 2: Common techniques used for the quantification of suspended particles in lakes.



If measured frequently enough, particle concentration data can thus be used to describe the fast processes of autochthonous particle formation in and allochthonous particle plumes to a lake as well as their distribution in the water column and throughout the lake basin. On the other hand, the concentration of particles does not seem to be an adequate parameter to characterize lakes varying in their trophic states, geochemical backgrounds, sizes, depths, etc.

2. Particle flux

Sediment trap measurements of the downward flux of particles in different lakes show annual mean values of 2–3 $\text{g} \cdot \text{m}^{-2}\text{d}^{-1}$ (Table 1). Higher fluxes (5–6 $\text{g} \cdot \text{m}^{-2}\text{d}^{-1}$) have been observed in some eutrophic lakes, where higher amounts of particles are produced during periods of increased productivity in the epilimnion (Lake Biel, Lake Greifen) [5,17]. Generally, particle flux data reflect seasonal changes of suspended material, whereby >70% of the suspensoids settle during spring/summer from May to September [6]. Periodic maxima of particle fluxes during this time indicate successions of phytoplankton blooms. These maximum values can be increased by synchronous plumes of allochthonous particles from spring melt water or torrential rains [5,7]. Short-term measurements (60 hours intervals) with time-sequencing sediment traps show that the duration of such increased pulses is usually short and seldom exceeds 2.5 days [2,19]. Determination of particle fluxes from traps just below the epilimnion and near the lake bottom, can show significant differences. They are caused by physical and biochemical recycling and transformation of particles outside or within a sediment trap.

Physical resuspension of already deposited material was observed in Lake Zug (South Basin) during a storm-event in November 1982, when a sediment trap exposed 9 m above ground measured a particle flux of 4.2 $\text{g} \cdot \text{m}^{-2}\text{d}^{-1}$ as compared to 1.7 $\text{g} \cdot \text{m}^{-2}\text{d}^{-1}$ (40%) for the trap set below the epilimnion [6]. Sinking Mn-oxides formed during the biochemical recycling of Mn (which comprises the release of Mn(II) from the sediment at the anoxic sediment/water interface and its subsequent oxidation to γ -MnOOH, catalyzed by microorganisms in the overlying water [15]) clearly increases particle fluxes in bottom traps of eutrophic lakes during periods of anoxia in the hypolimnion. An increased total particle flux (up to 7%) in the bottom traps and numerous Mn-oxide par-

ticles in both the water and trap samples indicate the influence of this short-cut recycling of Mn on sedimentation processes near the bottom during autumn and winter months in Lake Zurich [3].

On the other hand microbial degradation and grazing by filter-feeders within the traps, especially when exposed within or just below the epilimnion, may reduce trapped material during longer periods of exposure. During a 14-day trap experiment in Lake Greifen, more than half of the trapped material summed over 2-day intervals was degraded in the reference trap, which was continuously exposed during the experiment [17].

3. Sedimentation rates

Average annual sedimentation rates, displayed in Table 1, show surprisingly uniform values of about 0.4 $\text{cm} \cdot \text{y}^{-1}$ in most of the lakes listed. It has to be kept in mind, however, that lakes with large tributaries show annual rates, which depend on the position of the studied sediment core within the lake basin (e. g. near a delta or in more distal parts of the lake). Differences by up to an order of magnitude have been observed in some of the Swiss lakes, which are influenced by high debris-flow from rivers [5,7].

Average sedimentation rates are likewise significantly greater in those lakes, in which rates of productivity are superimposed by the input of high amounts of allochthonous material (e. g. Lake Biel [5]) or where frequent pulses of particle-loaded river water carry large quantities of allochthonous solids even to distal parts of a lake (e. g. Lake Brienz [7]). The rate of sedimentation, as determined by dating of sediment cores, describes the net amount of particles finally deposited on the lake floor. However, a sedimentation rate calculated from results from sediment trap material, which is susceptible to both recycling and biochemical alteration will usually be larger.

4. Composition of particles

As the formation and fate of suspended particles in lakes of temperate climates are seasonally variable, particle analysis of filtered samples and sediment trap material shows significant differences during the course of a year [6,18].

A dramatic increase in the number of diatom shells (*Stephanodiscus hantzschii*, *Asterionella* sp.) (Fig. 3) represents the first productivity peak of the year. Lakes with substantial par-

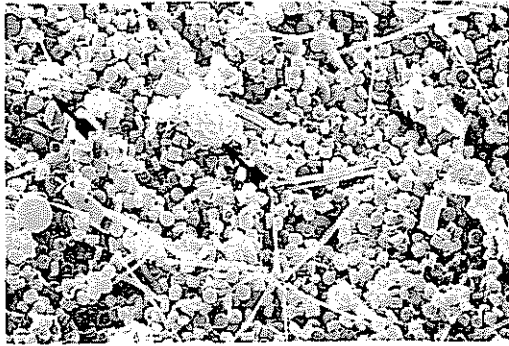


Fig. 3: A spring bloom of phytoplankton represented by a vast number of centric *Stephanodiscus hantzschii* and a few specimens of the pennate *Asterionella* sp. Clusters of diatoms (arrow) indicate fecal pellets from grazing zooplankton. Sediment trap material from the epilimnion of Lake Zug (South Basin), collected at 22 m water depth, April 20 - May 11, 1982. (bar = 100 μm)



Fig. 4: Autochthonous calcite crystal with residual holes of former chlorophyta nuclei. Water sample of pelagic Lake Constance, collected at 10 m water depth, May 23, 1981 (bar = 10 μm)

ticle input from rivers show simultaneous enrichment of allochthonous material in the samples (e.g. rock fragments, clay particles, quartz grains, etc.). In carbonate-saturated lakes sudden calcite crystal precipitation starts in early summer [20], when assimilation activity of the phytoplankton raises the pH and simultaneously decreases the reservoir of dissolved phosphorus in the lake, thereby no longer inhibiting crystal growth [16,20]. Newly formed calcite crystals reaching diameters of 40–60 μm , exhibit step-like morphology and inclusions of heterogeneous nuclei (shells of diatoms, chlorophyta as indicated by residual holes within calcites, Fig. 4). Epilimnetic calcite precipitation is characterized by high settling velocities (up to $100 \text{ m} \cdot \text{d}^{-1}$ [2]). The influence of these fast settling particles on the overall settling process is still unclear and subject of further studies. All the more as calculated settling velocities of total suspended matter (i. e. particle flux/particle concentration) are in the order of $1 \text{ m} \cdot \text{day}^{-1}$ [6]. Autochthonous calcite plumes may further be enhanced by summer phytoplankton productivity peaks during the summer, when large numbers of pennate diatoms and of calcareous greens (*Phacotus lenticularis*) are formed [3,20].

At the end of summer, when anoxic conditions are developed at the sediment/water interface in the deeper parts of mesotrophic and eutrophic lakes, biogenetic structures of Mn-oxide (*Metallogenium*) become more frequent in the suspended material (Fig. 5). These structures are present in particle samples from autumn until early spring. Then, during homothermic conditions, oxygen is mixed down to the deep hypolimnion and the first spring algal bloom starts a new annual cycle.

Suspended particles in lakes are part of a complex and interdependent environmental puzzle. Their quantitative and qualitative investigation helps to understand physical and geobiochemical processes which are essential for the function and the preservation of these natural systems.

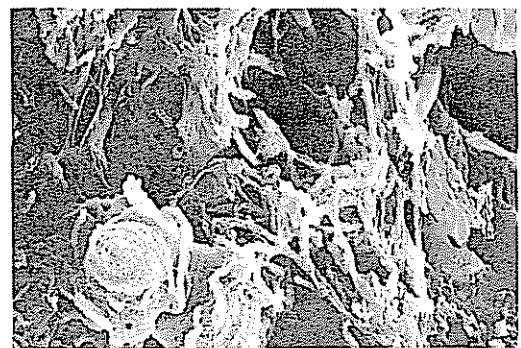


Fig. 5: Cabbage-like (left) and polygonal structures of Mn-oxides (*Metallogenium*) totally covering a 0.45 μm membrane filter of a hypolimnetic water sample of Lake Zurich, 130 m water depth, October 5, 1983. (bar = 10 μm)

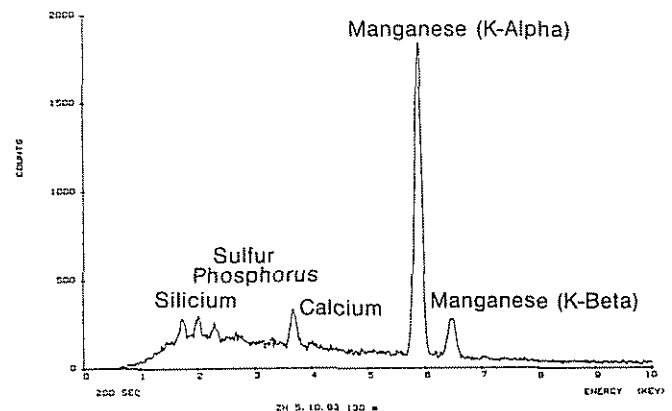


Fig. 6: Elemental composition of the Mn-oxides from Figure 5 (Analysis by SEM / EDS).

[1]–[12] see Table 1

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The Movement of a Tritium Pulse through Surface and Groundwaters in the Glatt Valley, Switzerland

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Present-day drinking water resources have become endangered by pollution due to anthropogenic activities. One of the parameters environmental scientists need to know in order to predict the fate and effect of pollutants in endangered surface and ground waters is the mean residence time of water, \bar{t}_w , in a given reservoir, or the mean transport velocity, \bar{v} , in a given ground water aquifer, stream, canal or lake. Unfortunately, these parameters are not always known very accurately. In ground water, which supplies most of our drinking water, \bar{t}_w is difficult to assess. In general, natural or artificial tracers could be used to determine \bar{t}_w or \bar{v} , but these often suffer from drawbacks, such as adsorption on particles, biological degradation, hydrolysis, photolysis, or gas exchange.

On December 13, 1983, an accidental release of tritium by an isotope processing plant resulted in a nearly optimal situa-

tion for measuring flow parameters in the watershed of the river Glatt (Fig. 1). Tritiated water is an almost ideal tracer because it is not microbially degraded and its transport is very little retarded by interactions with particles. EAWAG and EIR scientists responded by using an existing ground water test field, as well as other sampling stations, to follow the tritium pulse through the watershed. The continuous monitoring stations of the Swiss National River Surveillance Project (NADUF) helped to establish a mass balance. It thus became possible to characterize the flow of a conservative substance through a water treatment plant, and to validate a transport model for river water infiltration into ground water – derived previously from studies using EAWAG's test field – for a considerably larger area. The results of some previous studies using this test field in the lower Glatt River valley near Glattfelden, which was established by EAWAG in 1979 in order to understand the processes controlling the infiltration of polluted river water into an aquifer used as a drinking water reservoir, are described in [1-5].

Unfortunately, sampling did not start until 3-6 days following

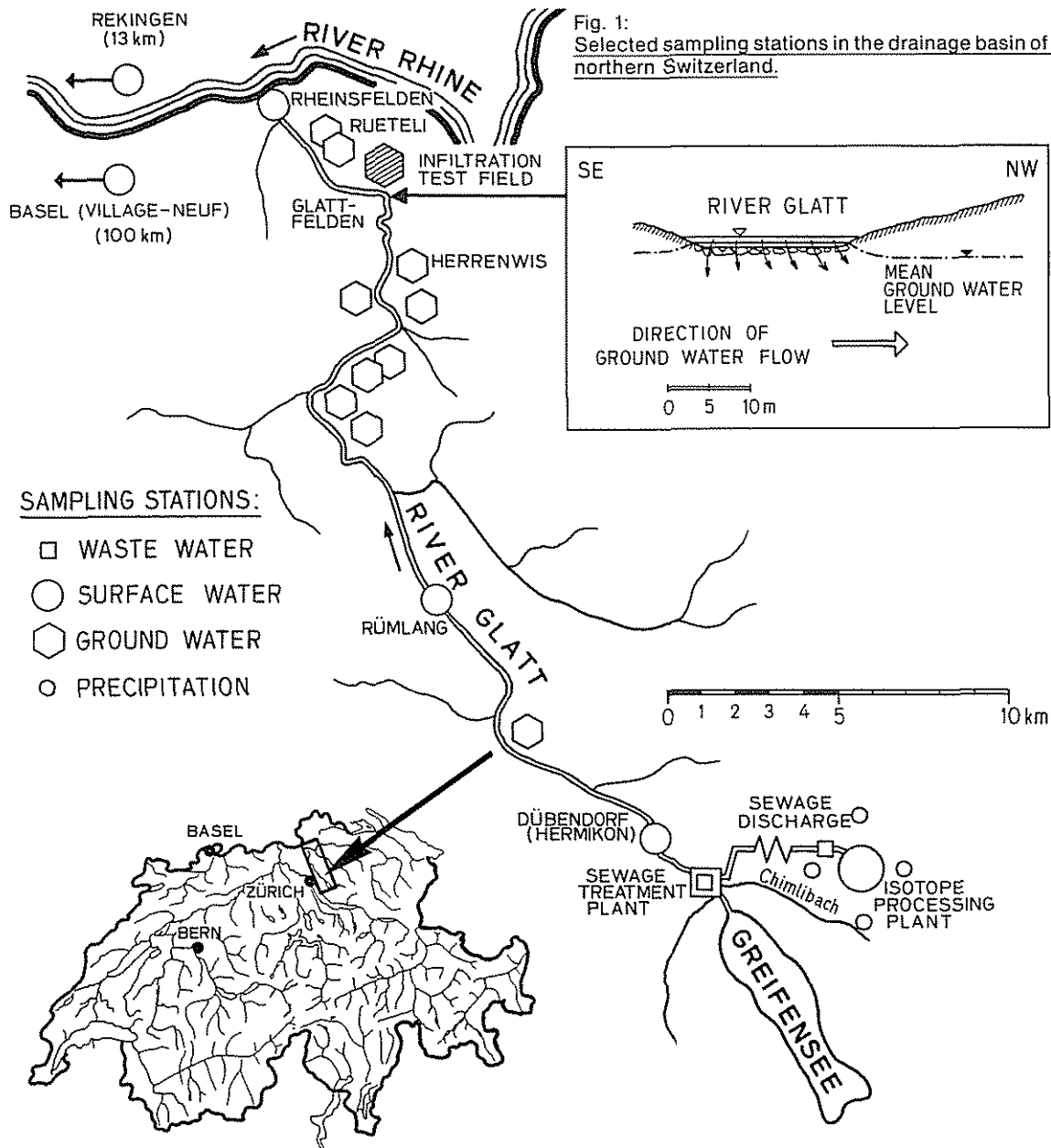


Fig. 1: Selected sampling stations in the drainage basin of the river Glatt in northern Switzerland.

the tritium release. Nonetheless, the 500 Ci pulse was strong enough to trace over more than 100 km for several months (Table 1).

The consequences and dose rates resulting from the accidental release were being dealt with by the Swiss National Accident Insurance Fund (SUVA), the Federal Health Department (BAG) and the Federal Commission for the Surveillance of Radioactivity in Switzerland (KUER). The dose rates were found to be insignificant. In this article we can concentrate on the scientific aspects of the accidental tritium release.

1. Memory effects of the environment after accidental releases of contaminants

Even though one might consider such a release a pulse, being quickly carried away from the injection point by surface waters, one has to bear in mind that such a pulse will be modulated by any reservoir the substance is flowing through. Removal processes to sediments and atmosphere as well as exchange processes with additional reservoirs coupled in parallel will also act on any chemical substance (Fig. 2), further delaying the response in the surface water downstream. This can lead to a memory effect of the whole system. Exactly this situation was observed following the accidental discharge of tritium described here.

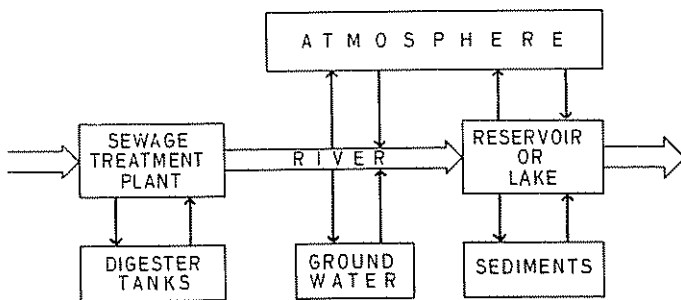


Fig. 2: Schematic of a typical situation after an accidental release of a contaminant into the environment.

The contaminant pulse will be modulated by a number of reservoirs coupled in series or in parallel to the main flow path.

Fig. 3 shows the general trend of the measured tritium concentrations: decreasing amplitudes and increasing time-lags of the response curves along the course of water flow. Peak concentrations in the inflow and outflow of the sewage treatment plant or in the Glatt river were, however, not measured (see Table 1 and Fig. 3), but rather had to be estimated.

The slow exchange with additional reservoirs delayed the response of the surface water systems. For example, even though the tritium release acted essentially as a pulse stimulus through the sewage treatment plant, the concentrations of tritium in flow-proportional weekly samples, taken from the Rhine River at Village-Neuf near Basel, were nearly constant between December 12–26, 1983 (Table 1 and Fig. 3). Additional reservoirs along the Rhine River caused this delayed response. Further examples will be discussed below.

A difference of about eight orders of magnitude was found between the highest concentration measured in the inflow to the sewage treatment plant and the background concentrations of the various monitoring stations (100–200 pCi/l), the latter of which are predominantly determined by the atmospheric bomb-tests conducted between the early 1950's and 1963.

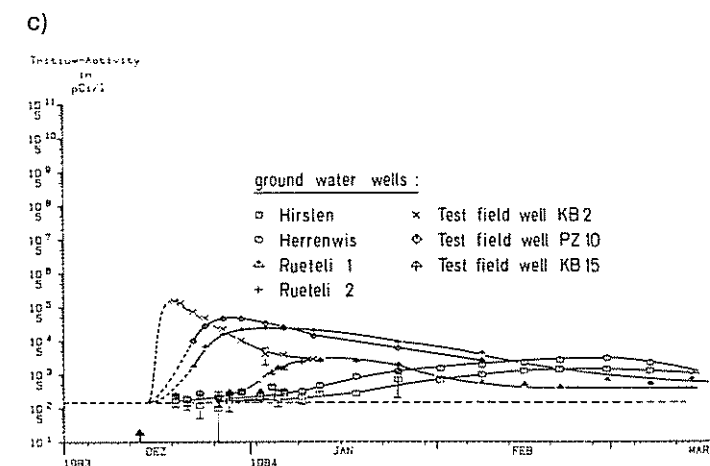
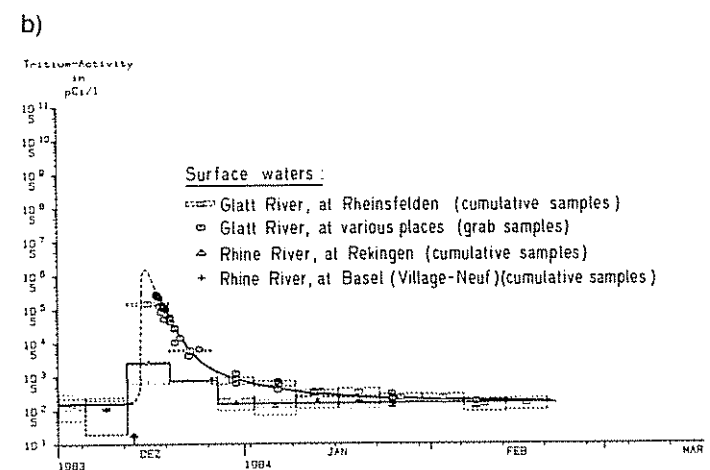
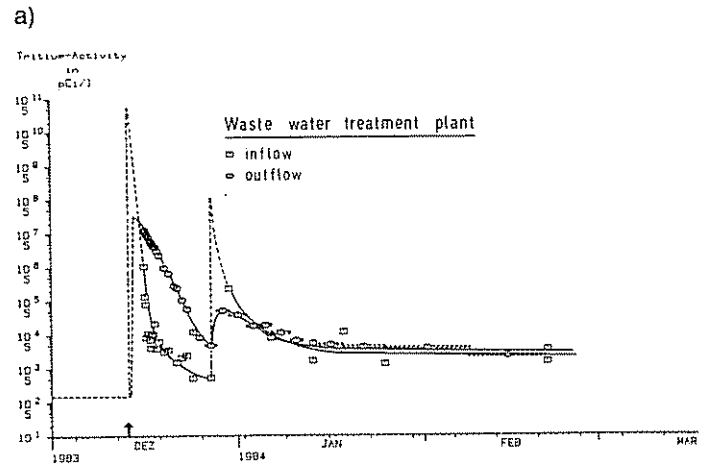


Fig. 3: Time dependence of the measured tritium concentrations in sewage discharge, surface and subsurface waters at the sampling stations shown in Fig. 1. Dotted curves are extrapolations based on the calculated total amounts of tritium discharged during the event, solid lines are fits through the data points. An additional small release of tritium of about 2 Ci by the isotope processing plant caused the increase in the tritium concentrations in the waters of the sewage treatment plant on December 26–27, 1983.

Table 1:

Estimated peak concentrations of tritium at selected sampling stations

	Q (m ³ /sec)	C _{max} (pCi/l)		Date of C _{max} in December 1983	
		measured	estimated ⁴⁾	measured	estimated
Sewage discharge of processing plant	1)	5.0 x 10 ¹⁰	5 x 10 ¹⁰	13. ⁵⁾	13.
Sewage treatment plant effluent	0.1	1.3 x 10 ⁷	5 x 10 ⁷	16.	14.
River Glatt at Hermikon	2	3.0 x 10 ⁵	2 x 10 ⁶	17.	14./15.
River Glatt at Rheinsfelden	4.5	1.5 x 10 ⁵	1 x 10 ⁶	12.-19. ³⁾	15.
River Rhine at Rekingen	340	2.5 x 10 ³	1 x 10 ⁴	12.-19. ³⁾	15.
River Rhine at Basel (Village Neuf)	720	7.5 x 10 ²	4 x 10 ³	12.-26. ³⁾	15.
Ground Water at Test Site (Well KB 2)	0.5 ²⁾	1.5 x 10 ⁵	2 x 10 ⁵	19.	18.

1) V ≈ 10 m³

2) Total infiltration flow rate [6]

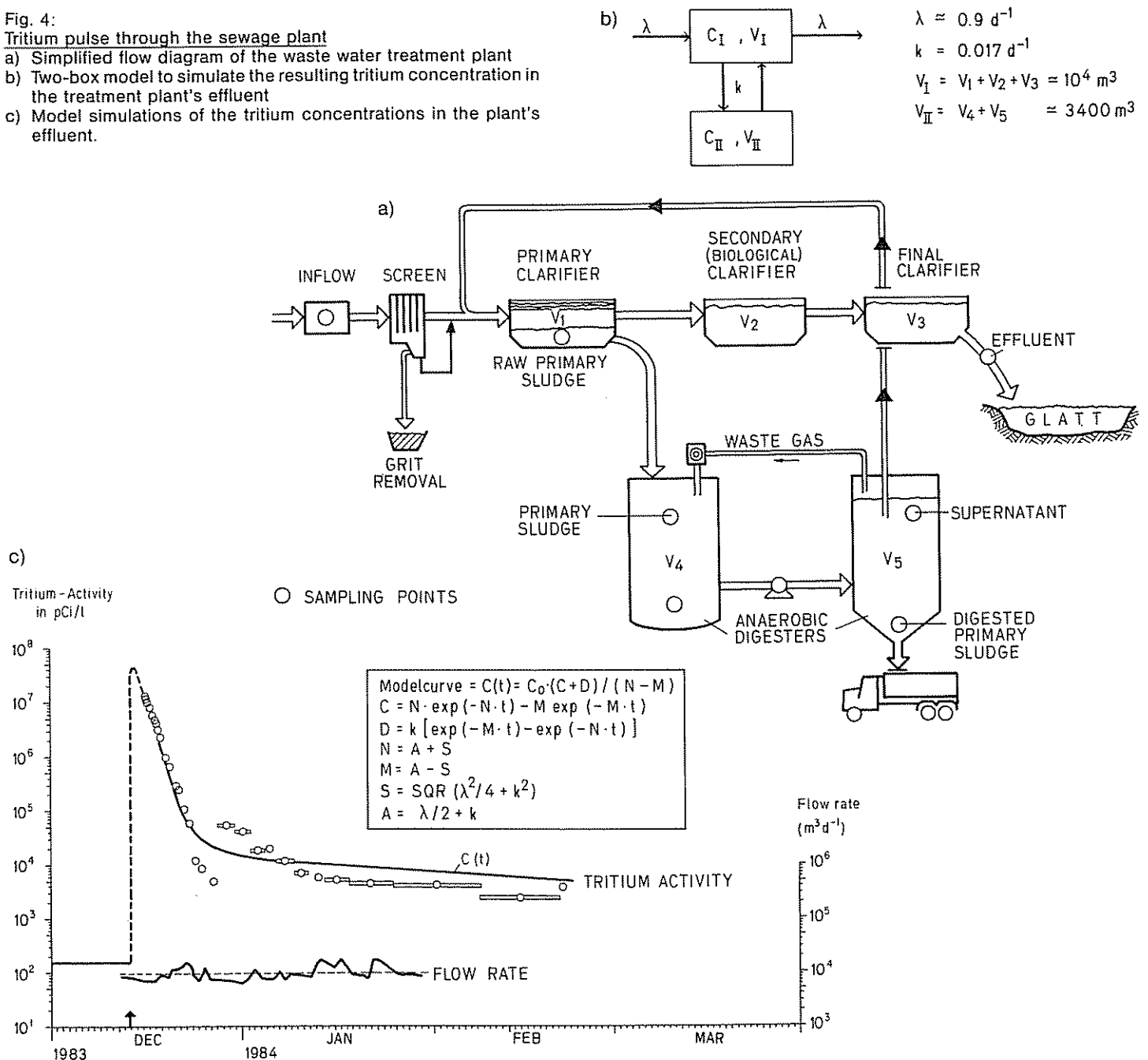
3) weekly average

4) The estimates are based on the measured total amount of tritiated water discharged by the Glatt River at Rheinsfelden of ca. 450 ± 50 Ci, the Rhine River at Rheinsfelden of 650 ± 150 Ci, Rhine River at Basel (Village-Neuf) of 400 ± 110 Ci, Rhine River at Rekingen of 410 ± Ci and measurements made by ⁵⁾ of 480 ± 10 Ci.

5) Personal communication from the isotope processing plant

Fig. 4: Tritium pulse through the sewage plant

- a) Simplified flow diagram of the waste water treatment plant
- b) Two-box model to simulate the resulting tritium concentration in the treatment plant's effluent
- c) Model simulations of the tritium concentrations in the plant's effluent.



2. Tritium pulse through the sewage treatment plant

The tritium release in the waste discharge of the isotope processing plant acted as a spike input to the sewage treatment plant. The expected residence time (\bar{t}_w) of the water in this treatment plant is about one day, which is equal to the total volume of 10^4 m^3 divided by the flow rate of ca. 10^4 (ranging from 6000 to 20 000) $\text{m}^3 \text{ day}^{-1}$ in the months of December and January. Indeed, the tritium concentrations in the effluent of the 3 clarifier basins of the plant (Fig. 4a) initially decreased exponentially with a residence time of about one day. This corresponds to the main pulse of tritium passing through the treatment plant. After approximately one week, the concentrations in the effluent decreased more slowly. The residence time of about 60 days during that period is accounted for by the slow release of tritium initially trapped by the two anaerobic digester tanks. Water in these two tanks is recycled into the treatment plant's primary clarifier basin with a flow rate which would fill the two tanks in about 60 days. A two-box model, which assumes constant flow rates, can adequately describe the water flow through the plant (Fig. 4b). The first reservoir in this simple model comprises the 3 clarifier basins, the second one consists of the two digester tanks in series. Deviations of up to a factor of two from the model curve are caused by two reasons: a) the irregular flow rates which caused a further dilution of the tritium input from the digester tanks during peak flow rates, and b) a second but smaller tritium input from the waste discharge of the isotope processing plant on December 26–27 (Figures 3a and 4c).

3. Tritium pulse through the river Glatt and the infiltration ground water

The concentration profile in the effluent of the sewage treatment plant acted as an input function to the Glatt River. Assuming a flow velocity of about 0.9 m/sec [2], the tritium pulse traveled from the treatment plant's outlet to Glattfelden and Rheinsfelden in 8–10 hours. In the latter area, the river flows on a glaciofluvial granular aquifer and infiltrates to the ground water [4].

The concentration profile of the quasi-conservative water tracer tritium in the river Glatt was assumed to act as a pulse stimulus input (i. e., a Dirac delta function) to the ground water. Mean residence times (\bar{t}_w) of 10 to 30 days in the infiltrating ground water with 26 to about 500 m flow distance (x) from the river, and approximate mean linear ground-water flow velocities (\bar{v}) of about 5 m/day were calculated from the breakthrough curves of Fig. 3. These values are in agreement with values determined earlier in the testfield [2, 4] and are typical for perialpine granular aquifers. The tritium concentrations in the ground water were simulated with a one-dimensional advection–dispersion model (Fig. 5). The freshly infiltrated water did not mix well with older ground water as was observed earlier in the testfield [4].

From the integration of the breakthrough response curves, we can obtain the ratio of total mass m divided by the flow rate Q (see Fig. 5). Since we know Q , we can calculate m . The value of Q was previously determined in [4] as $0.5 \text{ m}^3/\text{sec}$, and an interpretation of the geological cross-section given in [6] would lead to a somewhat smaller flow rate Q of about $0.2 \text{ m}^3/\text{sec}$. The value of m obtained this way is 25–50 Ci. We estimate therefore, that 5–10% of the tracer pulse of 500 Ci must have infiltrated to the ground water of the aquifer in the lower Glatt valley. This confirms the water loss of the river along the infiltration reach of 5–10% determined previously [3,4].

4. Conclusions

1) Accidental contaminant releases to the environment should be monitored closely, even if no person or ecosystem might be directly endangered. This is also true for releases of radioactive substances. If the monitoring is carried out

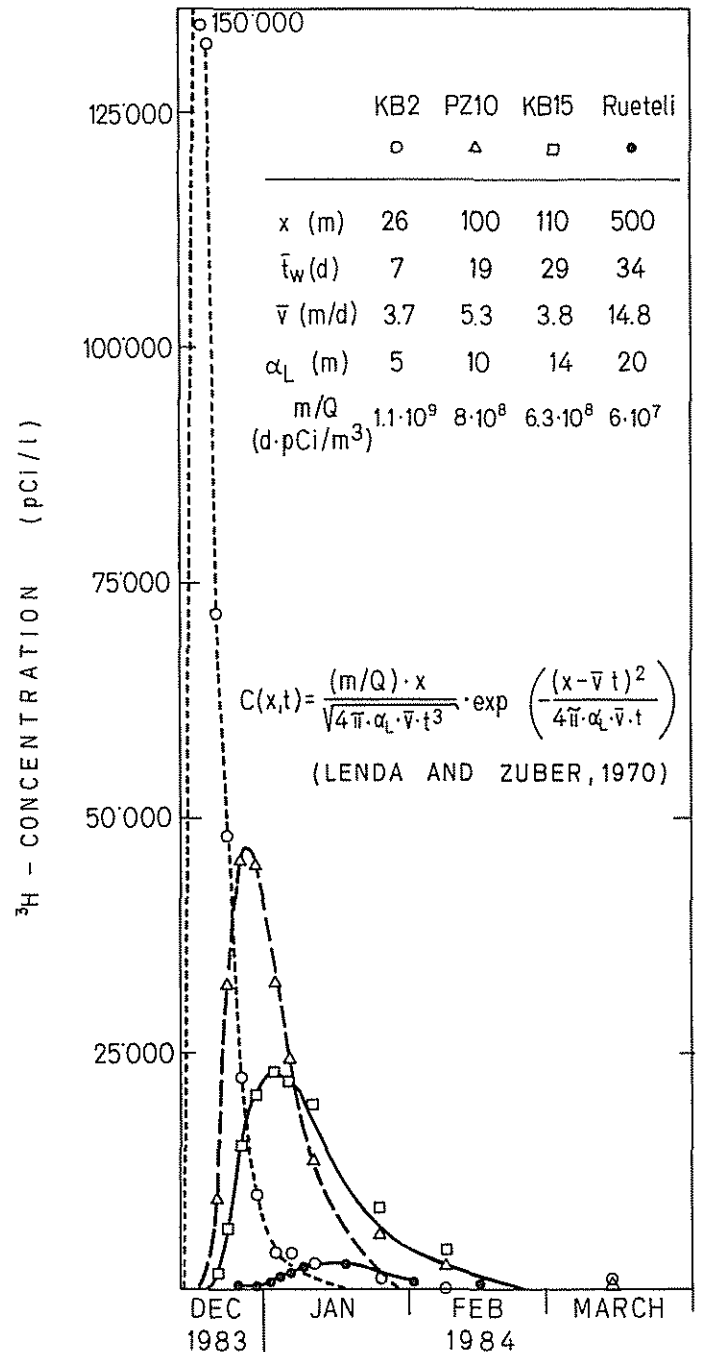


Fig. 5: Simulation of tritium concentration in the ground water. The measurements of the tritium concentrations in selected ground water wells are simulated by a one-dimensional advection–dispersion model using the given parameters. α_L = longitudinal dispersivity, m = total amount of tracer, Q = flow rate, $\bar{v} = x/\bar{t}_w$, with \bar{v} = mean linear ground water flow velocity, \bar{x} = mean infiltration distance, \bar{t}_w = mean ground water residence time [7].

properly, either the fate of this chemical substance or the geochemical processes controlling its dispersion in the environment can be studied, as was done here.

2) After a conservative chemical substance such as tritiated water reaches the surface waters, the affected waters exhibit a memory effect, i. e., concentrations in outflows of sewage treatment plants, rivers or reservoirs are modulated. The response curves in aquatic systems downstream from the point of injection of conservative substances, therefore, show timelags which depend on the volume of the reservoir involved and its exchange rate. Additional modulations would further result for non-conservative substances which could degrade or exchange with particles or with the atmosphere.

3) 5–10% of the tritium accidentally released into the Glatt River by an isotope processing plant was estimated to have infiltrated to the ground water along the course of the river. The concentrations of tritium in a number of ground water wells were easily measurable but still very low when compared with maximum permissible concentrations. The infiltrating tritium allowed validation of the experimentally derived transport model for river water infiltration into a glacio-fluvial granular aquifer for a considerably wider area.

Acknowledgements: The tritium analysis of the ground water samples of the test field were carried out by the Radiation Monitoring Department (SU) of EIR. The scientific interpretation of the accidental release was only possible thanks to the foresight of a number of people involved in the planning of a sampling strategy. O. Huber, H.R. Von Gunten, W. Stumm and H. Voelkle advised in various stages of the work.

[1] Giger, W., Schwarzenbach, R., Hoehn, E., Schellenberg, K., Schneider, J.K., Wasmer, H.R., Westall, J., Zobrist, J.: Das Verhalten organischer Wasserinhaltsstoffe bei der Grundwasserbildung und im Grundwasser. Gas – Wasser – Abwasser, 63 (9), 517–531, (1983).

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- [6] Freimoser, M., and Locher, Th.: Gedanken zur pleistozänen Landschaftsgeschichte im nördlichen Teil des Kantons Zürich, aufgrund hydrogeologischer Untersuchungen. Eclogae geol. Helv., 73 (1), 251–270 (1980).
- [7] Lenda, A., and Zuber, A.: Tracer dispersion in ground water experiments. Proc. of a Symposium, Vienna, March 9–13, 1970, IAEA – SM – 129/37, Vienna, 619–641, (1970).

The effect of Ozone on Waters containing Chlorine or Bromine
(p. 1–3)



Suspended Particles in Lakes (p. 3–6)



The movement of a Tritium Pulse through Surface and Groundwaters in the Glatt valley
(p. 6–11)



About the authors:

Dr. Werner Haag has been a research associate in the Chemistry Department of EAWAG since 1981. He received a B.S. degree in Chemistry from the Massachusetts Institute of Technology and a Ph. D. degree in Chemistry from the University of Tennessee, USA. His dissertation research was in the field of water chlorination kinetics.

Dr. sc. nat. Jürg Hoigné, assistant head of the Chemistry Department at EAWAG, received his Ph.D. degree in Physical Chemistry at the ETHZ, before specializing in the field of radiation chemistry by performing postdoctoral research in Brookhaven (USA) and the ETHZ. Following 7 years in applied industrial research, he has become a leading scientist in the field of ozone chemistry and in the kinetics of water treatment and environmental processes. As assistant professor of aquatic chemistry at the ETHZ, he lectures in water and wastewater technology and drinking water hygiene. A related review report coauthored with Heinz Bader appeared in the EAWAG-News Nr. 10 (Aug. 1979).

Dr. Michael Sturm is a geologist-sedimentologist with special interests in the formation and transformation of particles and sediments in the lacustrine environment. He is affiliated to EAWAG since 1977 and is a senior scientist of the Department of Multidisciplinary Limnological Research.

Dr. Eduard Hoehn is a hydrogeologist at EIR (Swiss Federal Institute for Reactor Research) and was a visiting scientist at Stanford University in 1980/1981. His research on the role of aquifers in the transport behavior of ground-water contaminants is a stimulation for researchers at EAWAG with whom he cooperates. (EAWAG publications 929, 939, 960, 967.)

Dr. Peter Santschi (see page 14) and his co-workers from left to right: *Alfred Lück, Klaus Farrenkothen, Esther Werth* and *Hans-Jürgen Hüppi*.



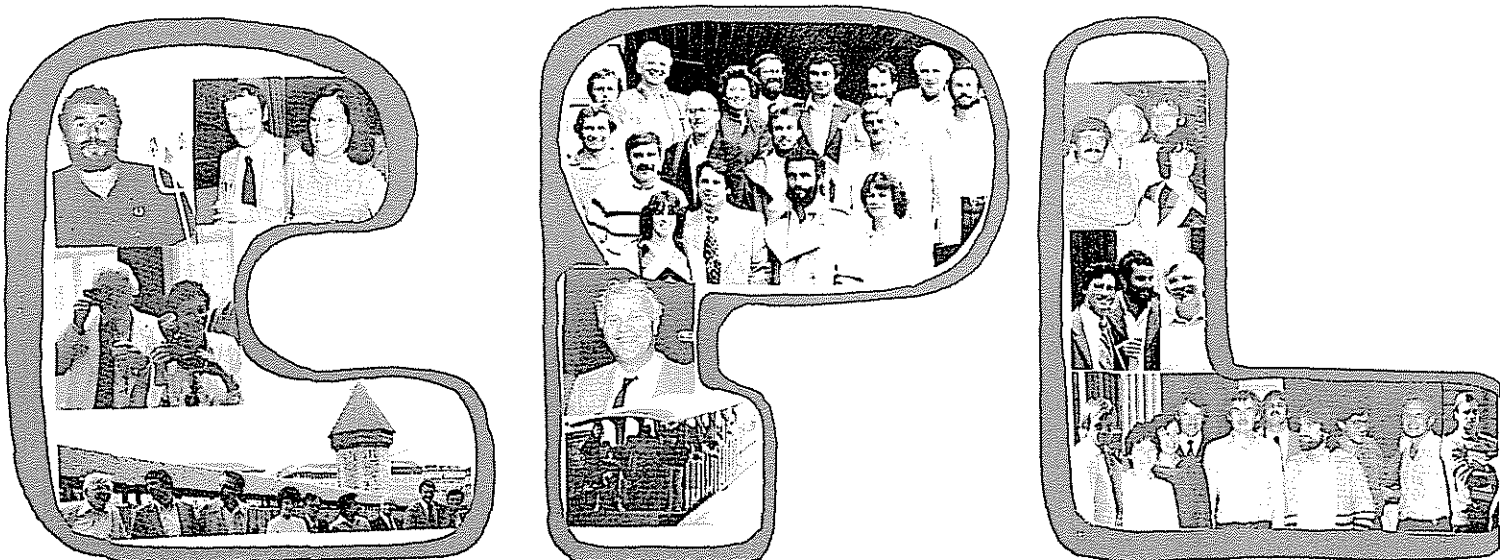
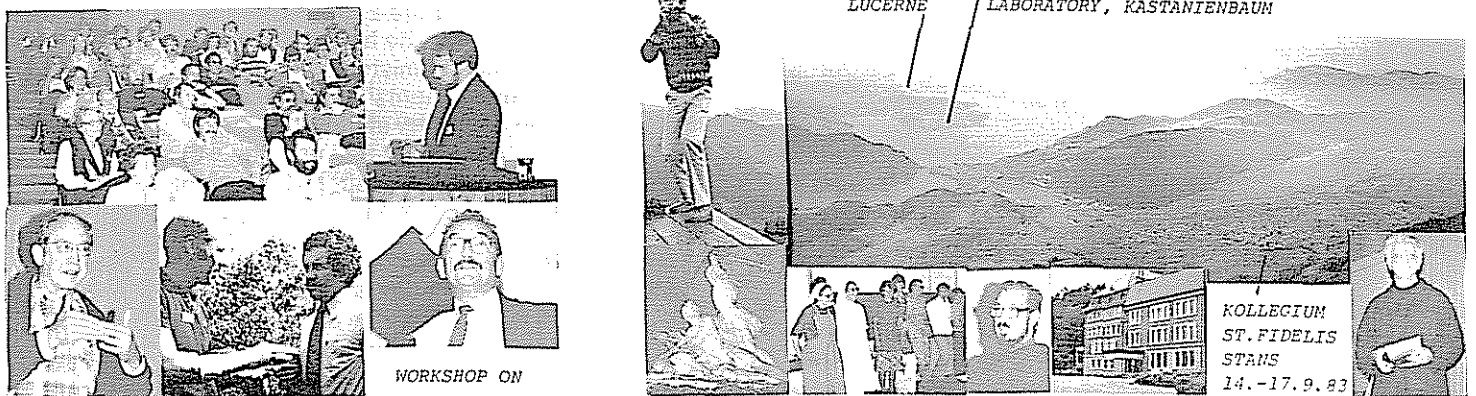
Workshop on «Chemical Processes in Lakes»

This workshop took place in September 1984 with nearly 100 participants in Kastanienbaum/Stans under the chairmanship of Professor *Werner Stumm*. Some of the highlights of the workshop are reflected in the photographs given below. As a result of this workshop a book entitled «Chemical Processes in Lakes», W. Stumm, ed., is in press by Wiley Interscience, New York.

The book considers the following topics:

- Redox Processes: Nutrient and Metal Interactions
 - Particle/Water and Sediment/Water Interactions
 - Chemical Perturbations of Lakes
- and has the following table of contents:
- Spatial and Temporal Distribution of Chemical Substances in Lakes: Modelling Concepts, *Dieter M. Imboden, René P. Schwarzenbach*, EAWAG, Switzerland
 - Conceptual Models for Transport at a Redox Boundary, *William Davison*, Freshwater Biological Association, Ambleside, UK
 - Aqueous Surface Chemistry: Assessment of Adsorption Characteristics of Organic Solutes by Electrochemical Methods, *Božena Čosović*, Budjer Bošković Institute, Center for Marine Research, Zagreb, Yugoslavia
 - Strategies for Microbial Resistance to Heavy Metals, *John M. Wood, Hong Kang Wang*, Gray Freshwaters Biological Institute, Univ. of Minnesota, Navarre, USA
 - Carbon Isotopes and Productivity in Lacustrine and Marine Environments, *Judith McKenzie*, Institute of Geology ETH Zürich, Switzerland

- Redox-related Geochemistry in Lakes; Alkali Metals, Alkaline-Earths Elements, and ^{137}Cs , *Edward R. Sholkovitz*, Woods Hole Oceanographic Institute, Woods Hole, USA
- Mechanisms Controlling the Sedimentation Sequence of Various Elements in Prealpine Lakes, *Hans-Henning Stabel*, University of Constance, Fed. Rep. of Germany
- The Pavin Crater Lake, *Jean-Marie Martin*, Ecole Normale Supérieure Paris, France
- Phosphate Interactions at the Sediment/Water Interface, *Peter Baccini*, University of Neuchâtel and EAWAG, Switzerland
- The Influence of Coagulation and Sedimentation on the Fate of Particles, Associated Pollutants, and Nutrients in Lakes, *Charles O'Melia*, The John Hopkins University, Baltimore, USA
- The Coupling of Elemental Cycles by Organisms: Evidence from Whole Lake Chemical Perturbations, *David W. Schindler*, Freshwater Institute, Winnipeg, Canada
- The Geobiological Cycle of Trace Elements in Aquatic Systems: Redfield Revisited, *François M. M. Morel, Robert J.M. Hudson*, M.I.T., Cambridge, USA
- Metal Transfer Mechanisms in Lakes: the Role of Settling Particles, *Laura Sigg*, EAWAG, Switzerland
- Acidification of Aquatic and Terrestrial Systems, *Jerald L. Schnoor*, University of Iowa, USA, *Werner Stumm*, EAWAG, Switzerland
- The Chemistry of Bog waters, *Evilve Gorham, Steven J. Eisenreich, Jesse Ford, Mary V. Santelmann*, University of Minnesota, Minneapolis, USA
- Lake Restoration, *René Gächter*, EAWAG, Switzerland
- Kinetics of Chemical Processes of Importance in Lacustrine Environments, *James J. Morgan, Alan Stone*, California Institute of Technology, Pasadena USA



On a lighter tone here are a few «choice» excerpts from the experts at the workshop

«... simple and complex ...»

«useful but in error ...»

«The different values are not the same.»

«Sometimes we have correlations. Sometimes we don't.»

«The effects can be significant or negligible.»

«The point is that the constants are not really constant.»

«My calculations are right ... and I suppose so are your calculations ...»

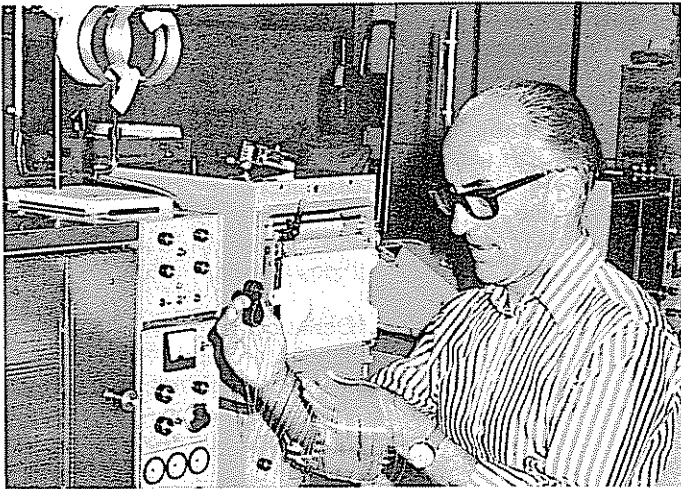
«We're quite sure of this point, but we don't know what it represents.»

«... except for the biology ...»

(Collected by Dr. Patricia Colberg, EAWAG)

News about EAWAG Collaborators

Dr. Kurt Grob, expert on the application of capillary gas chromatography for the separation of organic pollutants, has been promoted to Professor of the Swiss Federal Institute of Technology, recognizing his merits in the art of teaching and developments in capillary gas chromatography.



The Association of Environmental Engineering Professors (AEPP) recently awarded «The Outstanding Publication Award» to Professors Werner Stumm and James J. Morgan



for their 1962 article «Chemical Aspects of Coagulation» which was published in the Journal of the American Water Works Association. This is the first such award. An outstanding publication will be chosen each year in the future. Criteria for the selected paper must be on «significant research and of educational value» and one which has «stood the test of time». James Morgan is Professor of Environmental Engineering Science at the California Institute of Technology, Pasadena, USA. Werner Stumm is shown receiving the award from Professor Joseph H. Sherrard, a member of AEPP who was at EAWAG on a study research leave from Virginia Polytechnic Institute and State University during 1983–1984.



The Faculty of Natural Sciences has awarded the ETHZ silver medal to Johannes Stähelin for his dissertation on «Ozone decay in water: Kinetics of the initiation by hydroxide ions and hydrogen peroxide and the Subsequent Reactions of the Hydroxyl and Peroxide Radicals» (in German). This thesis research was carried out at EAWAG under the supervision of Dr. Jürg Hoigné. Dr. Stähelin is now at the University of Colorado, Boulder (USA), for postdoctoral studies on atmospheric chemistry. In 1983, this award was given to Dr. Erich Staub, now working in the Swiss Environmental Protection Agency.



This year's Otto Jaag Prize for the most outstanding thesis in the field of water resources and water pollution control was awarded to Andreas Frutiger for his thesis entitled: «Studies on the Ecology of the Predacious Stonefly *Dinocras cephalotes* Curt. (Plecoptera: Perlidae) in a pre-alpine Swiss river» (in German). In 1980, this award was given for Ph. D. research conducted at EAWAG by Dr. Laura Sigg, Head of Analytical Chemistry Section, EAWAG, and in 1981 to Hansjörg Fricker, presently in the Department of Zoology, University of Alberta, Edmonton, Canada.



Dr. René Schwarzenbach has been promoted to Privatdocent* for Environmental Sciences at the Federal Institute of Technology (ETHZ, Dept. X). The title of his habilitation thesis is: «Assessing the Behavior and Fate of Hydrophobic Organic Compounds in the Aquatic Environment – General Concepts and Case Studies emphasizing Volatile Halogenated Hydrocarbons».

Dr. R. Schwarzenbach joined the Chemistry Department of EAWAG in 1977 after two years as a visiting research scientist at the Woods Hole Oceanographic Institution, Massachusetts, USA. In autumn 1983 he became an associate head of the Department of Multidisciplinary Lake Research (MLF) at EAWAG's research laboratory in Kastanienbaum on Lake Lucerne.

* «Privatdocent» is an academic title used by many universities in continental Europe. It is in many regards equivalent to an Assistant Professor in the USA. In order to become a «Privatdocent» a scholar writes a «habilitation thesis», which is like a second doctoral thesis at a higher scientific level. Upon acceptance of the thesis and after passing the examinations, the candidate acquires the «venia legendi», the right to give courses at the University.

As of September *Dr. Peter Santschi*, previously a senior research associate at the Lamont-Doherty Geological Observatory of the Columbia University (USA), became head of EAWAG's Section for Radioactivity and Isotopes succeeding *Dr. Maria M. Bezzegh*. His main research interests are geochemical processes in natural waters, using both natural and artificial radioisotopes and the fate of trace contaminants in lakes and oceans.

Dr. Santschi was also promoted to Privatdocent at the Swiss Federal Institute of Technology in the field of isotope geochemistry. His habilitation thesis was entitled «Radioisotopes as Tracers of Sediment-Water Interactions in Controlled Experimental Ecosystems: Case Studies». In this edition of EAWAG-News, *Santschi* and his co-workers *K. Farrenkothen*, *H. Hüppi*, *A. Lück* and *Esther Werth* and *Dr. E. Höhn*, from the Swiss Federal Institute of Reactor Research (EIR), describe the use of tritium as a tracer of water residence time and of river water infiltration into a groundwater system.

Since 1980, the Institute for Water Pollution Control (IGW) and the Institute for Hydraulics and Water Resources Management (IHW) of the Swiss Federal Institute of Technology (ETH) have been offering a **postgraduate course in Sanitary Engineering and Water Pollution Control**. It is one of the eight accredited postgraduate courses of the ETH. A fifth course will begin at the end of October 1984.



Participants of the fourth postgraduate course:
From left to right, *first row*: Christian Voegli, Basil Stotz, *Dr. Jürg Heldstab*, Martin Adämmer, Rolf Knoll; *second row*: Hanspeter Zeller, Verena Sturzenegger, Bettina Schumpelick, Christian Schaffner; *third row*: Bernhard Wehrli, Annette Johnson, Ulrich Sieber, Alfred Wüest, Heinrich Senn, Bo Gunnar Swensson, Colin Anthony Mason.

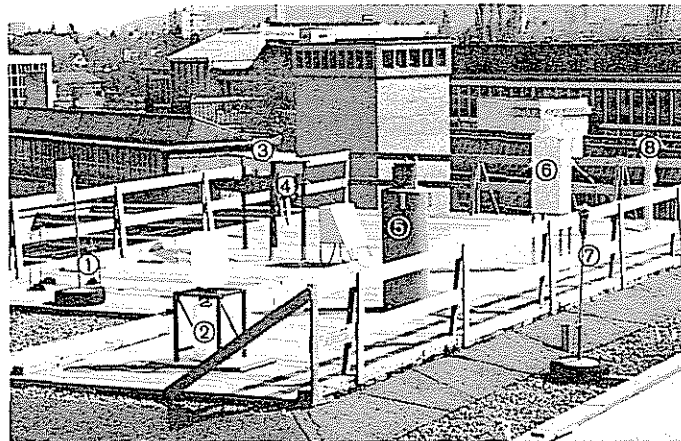
The postgraduate course is designed for biologists, chemists, physicists and engineers holding a degree equivalent to an ETH diploma. Practical experience in the field of environmental science and technology is desirable. A good reading knowledge of German and facility with applied mathematics are required.

The course, which runs 32 weeks, is divided into 3 parts. The first nine weeks are reserved for an introduction into basic Sanitary Engineering. Engineers holding a diploma in Civil and Rural Engineering need not participate in the first section. The following two parts of 10 and 13 weeks each, respectively, last from the beginning of January until mid-July with a break of three weeks in the spring. The course may also be attended over a period of two years. The number of participants is limited to 15.

Interested candidates should contact *Dr. Markus Boller*, EAWAG, Secretary of the postgraduate course (preferably before July 1984).

EAWAG has begun a research program on the **chemistry of atmospheric deposition** (dry deposition, rain, fog and aerosols) and its impact on aquatic and terrestrial ecosystems.

The research, sponsored by the Swiss National Science Foundation's National Program on «Air Quality and Air Resources», is directed jointly by *Dr. Jürg Zobrist* (Methods of collection), *Dr. Laura Sigg* (Inorganic chemistry) and *Dr. Walter Giger* (Organic chemistry).



The photograph below shows the following apparatus: rain gauge (1), bulk collector (composite samples, large volumes, 2), bulk collector (glass, 3; plastic, 4; for single events), wet-only collector (5), wet-only collector (6), sensor for wet-only collector 5 (7), fog collector (to be installed, 8).

In 1983, the following **guest research scientists** visited the respective departments of EAWAG:

Ahel Marijan, Dipl. Chem., Institut Rudjer Boskovic, Zagreb, Jugoslawien (Oct. 82 – Sept. 83);

Chin-Feng Pao-Kuo, Prof., Fairleigh-Dickinson Univ., Teaneck, N.J., USA (June – Aug. 83);

Christensen Erik, Prof., Dept. of Civil Eng. der Univ. of Wisconsin, Milwaukee, WI, USA (June – Aug. 83);

Goncalves Maria de Lurdes, Prof., Centro de Quimica Estrutural, Instituto Superior Tecnico, Lisboa, Portugal (July – Aug. 83);

Murray James, Prof., Dept. of Oceanography der Univ. of Washington, Seattle, Wash., USA (Jan. – Sept. 83);

Schnoor Jerald, Prof., Dept. of Civil and Environmental Engineering, Univ. of Iowa, Iowa City, USA (March – June 83);

Sharefkin Mark, Dr., Resources for the Future, Washington D.C., USA (Dez. 83 – March 84);

Sherrard Joseph, Prof., Dept. of Civil Eng., Virginia Polytechnic Inst. and State University, Blacksburg, Virginia, USA (Sept. 83 – March 84);

Wan Guojian, Dr. sc. techn., Institute of Geochemistry, Academia Sinica, Guiyang, Province Guizhou, China (July 83 – July 84);

Zengh Shu-Di, The Third Designing Inst. of the Ministry of Chemical Industry, Shanghai, China (Jan. 82 – Aug. 83).

As in previous years, the **course on «Hydraulics and Pollution of Ground Water»** was held at the ETH Höggerberg from March 12–16 (1984). The lecturers were: *Prof. Dr. Jacob Bear* (Haifa, Israel), *Prof. Dr. Ghislain de Marsily* (Fontainebleau, France) and *Dr. René Schwarzenbach* (EAWAG, Switzerland).

The **Third International Waste Treatment and Utilization Symposium** (IWTUS-3) is being organized by EAWAG and will be held in Frauenfeld in northeastern Switzerland from 27–31 August 1984. The symposium proceedings will be published by Pergamon Press. For further information contact either *Prof. G. Hamer* or *Dr. J. D. Bryers* of the Technical Biology Department at EAWAG. An application form for symposium participation can be found on page 15. Registration fees include accommodation, meals, excursions and a copy of the proceedings.

EAWAG-Publications

Corrections and supplementary data

937
Haag W.R. Hoigné, J.: *Ozonation of Water Containing Chlorine or Chloramines*. *Water Research*, 17, 1397-1402 (1983).

943
Jones, D.M., Douglas, A.G., Taylor, J., Giger, W., Schaffner, C.: *The Recognition of Biodegraded Petroleumderived Aromatic Hydrocarbons in Recent Marine Sediments*. *Marine Poll. Bull.* 14, 103-108 (1983).

947
Grob, K.: *An Attempt to Extend Ordinary Capillary Gas Chromatography to Supercritical Fluid Chromatography (SFC)*. *J. HRC & CC* 6, 178-184 (1983).

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Stumm, W., Schwarzenbach, R., Sigg, Laura: *From Environmental Analytical Chemistry to Ecotoxicology - a Plea for More Concepts and Less Monitoring and Testing*. *Angew. Chemie, Int. Ed.*, 22, 380-389 (1983).

953
Bundi, U.: *Stand und Perspektiven der Gewässerschutzforschung in der Schweiz*. *Schweiz. Ing. & Architekt* 101, 629-632 (1983).

New Publications

954
Koch, M., Dolfing, J., Wuhrmann, K., Zehnder, A.J.B.: *Pathways of Propionate Degradation by Enriched Methanogenic Cultures*. *Appl. & Env. Microbiol.* 45, 1411-1414 (1983).

955
Stumm, W., Furrer, G., Kunz, B.: *The Role of Surface Coordination in Precipitation (Heterogenous Nucleation) and Dissolution of Mineral Phases*. *Croatica Chemica Acta* 56, 585-603 (1983).

956
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