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CHERNOBYL RADIONUCLIDES IN THE ENVIRONMENT:

Tracers for the tight coupling of atmospheric, terrestrial and aquatic geochemical processes

Peter H. Santschi

I. INTRODUCTION

Radioactive fallout from the burning Chernobyl reactor was measured during May and June 1986 all over the northern hemisphere, even though, at times, activities were at extremely low levels. The attention given in each country to the radioactivity measurements as a basis for estimating dose rates to man, and the relative ease of measurement of radionuclides, allowed scientists to follow the journey of the radioactive cloud around the world. The accidental release of radioactivity provided a striking demonstration of the fact that all trace contaminants, radioactive and non-radioactive, can be transported within days from one country to another via the atmospheric and aquatic conveyor belts. The radioactive fallout from the reactor accident in Chernobyl provided a pulsed release to the environment; as such, this pulse presented a good opportunity to study transport processes in atmospheric, terrestrial and aquatic reservoirs, as pulse inputs of radionuclides to the environment can be used to study physical (e.g., ^{3}H), chemical (e.g., ^{54}Mn) or biological (e.g., ^{14}C) or hydrological (e.g., ^{3}H [1]) processes operating in a particular system. Understandably though, most of the attention was given to the dose rate aspect of the accident, i.e., the transfer of Chernobyl radionuclides to man and relatively little to the geochemical behavior of the nuclides.

The purpose of this article is threefold:
1) to demonstrate, using selected radionuclides from the Chernobyl fallout as tracers, the tight coupling between the movement of water in the atmospheric, terrestrial and aquatic reservoirs in the Zurich area ;
2) to show which processes influence the rates of migration of radionuclides in the environment and transfer to man (fig. 1), and
3) to indicate the extent to which the information on the Chernobyl radionuclide pulse agrees with what has been learned from previous studies of bomb fallout nuclides and releases from previous reactor accidents. While some of the radioactivity measurements reported here were also made by a number of other groups, the radionuclide mobility studies in atmospheric and aquatic systems, carried out by research teams of the Atmospheric Physics Section of ETH, the Federal Reactor Institute (EIR), the Inorganic Chemistry Institute of the University of Berne, and of EAWAG, are unique.

2. METHODS

The gamma-ray emitting Chernobyl radionuclides were determined on solid state detectors (Ge-Li, and high-purity-Germanium) in samples from surface, drinking and groundwaters, sewage treatment plants, air filters, total precipitation, dry fallout, sedimenting particles from Lake Zürich caught in sediment traps, fish, grass, milk, salads and vegetables. In selected samples, beta-ray emitting ^{90}Sr and ^{89}Sr activities were determined as well [2]. Detector geometries were calibrated with standards of the appropriate radionuclide in order to circumvent summation corrections.

The deposition rate was measured in special collectors for total precipitation and for dry deposition, either on consecutive days or as the total over the fall-out period. Dry fallout was collected in pans filled with a thin layer of distilled water, open to the atmosphere only during times of no precipitation. Because surface water concentrations of ^{137}Cs soon became unmeasurably small with direct counting techniques on 1 liter samples, ^{137}Cs had to be preconcentrated from 1-30 liters by ion exchange before measurement.

3. CHEMICAL FORM OF THE NUCLIDES IN THE ENVIRONMENT

The composition of radionuclides found in the radioactive Chernobyl cloud in Switzerland was determined by the following factors (fig. 1):
a) The asymmetrical fission yield of ^{235}U which favors nuclides with neutron numbers near the "magical" numbers 50 and 82;

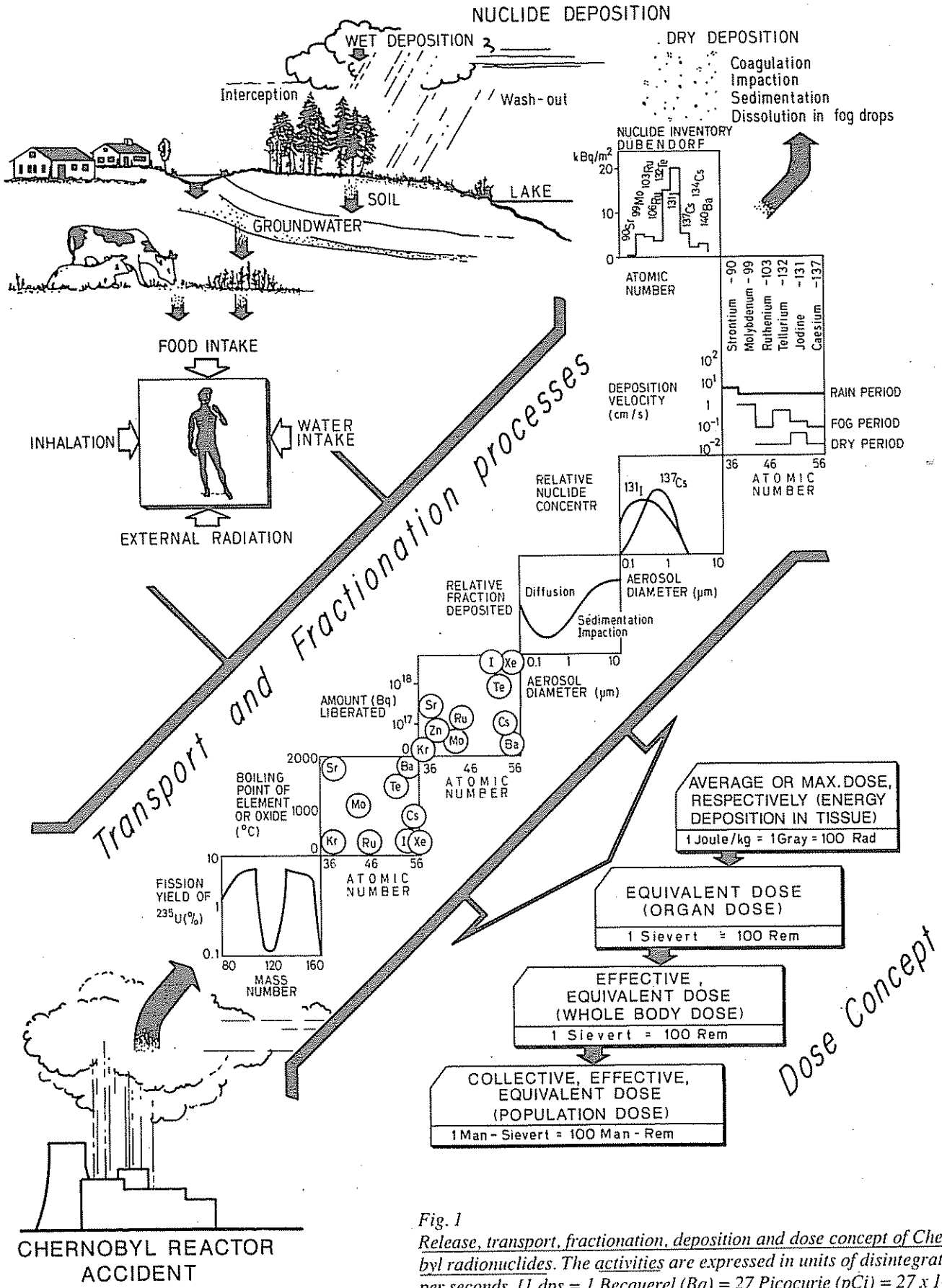

Fig. 1
Release, transport, fractionation, deposition and dose concept of Chernobyl radionuclides. The activities are expressed in units of disintegrations per seconds [1 dps = 1 Becquerel (Bq) = 27 Picocurie (pCi) = 27 x 10^{-12} Curie (Ci)]. The radiation exposure measured in air [unit: 1 Roentgen = 1.6 x 10^{12} ion pairs formed per g air] can be detected in an electrical field from the measurement of electrical currents. Ion pairs and radicals formed by an absorbed dose [1 joule / kg =1 Gray = 100 Rad] are responsible for mutagenic and cancerogenic effects.
Dose concept: An equivalent dose can be calculated for organs, whole body and for whole populations for present and, in case of long-lived nuclides, for future generations. In order to calculate an equivalent dose to man, one multiplies the number of Grays

b) the relative volatility of the elements or their oxides at 1500° C, the burning temperature of the reactor graphite [4], and
c)by fractionation and deposition processes in air.
Based on thermodynamic reasoning, the following chemical forms or oxidation states of selected Chernobyl radionuclides are likely: Cs^{+}, Ba^{2+}, La(III), Ce(IV), Nb(V), MoO_4^{2-}, TcO^{4-}, $HTeO^{4-}$. For ^{131}I and ^{103}Ru, for which it is also possible to postulate metastable forms, the following species or oxidation states are possible: $RuO4^{2-}$, Ru(III), I_2, CH_3I, I^- and IO_3^-. The speciation of the radionuclides was partially established in the burning reactor by the chemical and physical conditions there, which allowed the production of the volatile species I_2 and RuO_4. These proposed species or oxidation states are consistent with:
1) the observed behavior of the Chernobyl radionuclides in laboratory extractions by ion exchange and activated charcoal columns and by precipitation reactions; and
2) the observed nuclide mobility in the environment: uptake by and washout from soils and plants, and the groundwater infiltration behavior.

4. RESULTS AND DISCUSSION

4.1 Washout of the atmosphere by rain, fog and dew as the determining factor in the deposition of the Chernobyl radionuclides.

The concentrations of selected radionuclides collected in Dübendorf in air filters (Fig. 2), were similar to those in the rest of Switzerland [2], France and Germany, and only a little less than those measured in Sweden [3]. The deposition rates were, however, very different in different parts of Switzerland and in Western Europe. This was mainly caused by the unequal rainfall during the transit of the radioactive cloud. Nuclide deposition in Switzerland was particularly high near Lake Constance and in southern parts of the Canton of

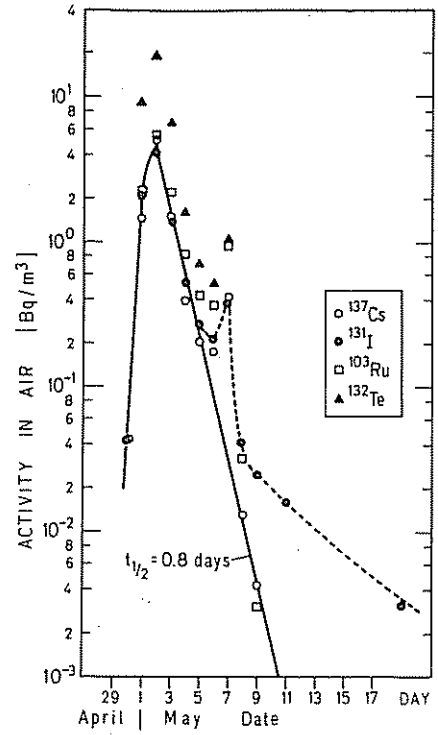

Fig. 2
Radionuclide activities in air filter samples in Dübendorf collected with a high volume sampler at 960 m^3d^{-1} flow rate (NABEL-Station).

Tessin (e.g., over Lake Lugano). The high efficiency of water droplets in rain and fog to transfer contaminants in the air to the ground is demonstrated by our measurements.
Table 1 shows the deposition rates and deposition velocities of selected radionuclides during periods of rainfall, fog and dry weather calculated from the respective activities in our atmospheric collectors. Deposition rates evaluated from activities in rain collectors of different geometries and from maximum activities in grass samples generally agreed to within 20 %. These measurements demonstrate the high deposition efficiency of rain events. These events accounted for 70-80% of the total deposition for $^{137,134}Cs$, $^{103,106}Ru$, ^{131}I and ^{132}Te, while the dry fallout, which was measured separately, only accounted for 20-25%. Furthermore, by calculating the ratio of the measured deposition rate (r_D, [kBqm^2]) to the integrated air concentration (c_{air}) on that day, deposition velocities (v_D) for the different radionuclides could be calculated as follows: $v_D = (r_D / \int c_{air} \cdot dt)$ [cm/s].
The calculation of these deposition velocities is particularly important as they can serve as an estimate for the deposition velocities of other contaminants in the air under similar atmospheric conditions. These calculations showed that the rain event of April 30, 1986, which lasted intermittently over a period of 12 hours, washed out the nuclides in the air at a velocity of about 3 cm/s. The average deposition velocity during the time, accounting for 90% of the nuclide deposition, was, however, only 0.4 cm/s. This is due to the much smaller deposition velocities produced during dry weather periods. Nonetheless, during fog periods, deposition velocities increased by about five fold as compared with dry weather.
The calculated deposition velocities are of the same order of magnitude as those estimated during the 1950's for the fission products of bomb fallout and for the nuclides from the reactor accident of Windscale in England in 1957. In the latter case, overall deposition velocities ranged between 0.1 - 0.3 cm/s, depending on the nuclide [5].
The deposition velocities calculated here for Chernobyl radionuclides during different weather conditions are, however, more detailed than what has been reported before.
The average scavenging ratio by rain droplets, S, is calculated from

$$S = r \cdot c_{rain}/c_{air}$$

with r = density of air (1.2 kg/m^3),and c_{rain} (Bq/kg) and c_{air} (Bq/m^3) the concentrationsof radionuclides in rain water and air, respectively. The average S, for different nuclides scavenged during the rain event on April 30, 1986, was similar for all nuclides at a value of 400. This is close to the value expected for continental aerosols [6] of an average diameter of 0.5 to 1µm [2]. This agreement should allow the calculation of deposition rates for other atmospheric trace contaminants which are associated with aerosols of similar sizes and which can be transported over distances of hundreds to thousands of kilometers over a continent.

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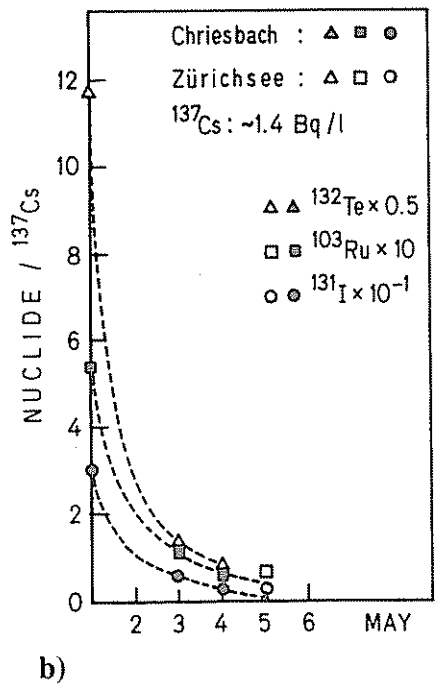
with a quality factor (QF) which considers the different linear energy transfer for different types of nuclear radiation (QF = 1 for beta- and gamma-rays, 10-20 for alpha- and neutron-rays.) The unit is the Sievert (Sv) = 100 Rem. In order to calculate the Effective Equivalent Dose, one sums over all organ doses, weighted by a factor which considers the different sensitivities to radiation damage of the different organs. According to international guidelines, one considers governmental activities to start at annual dose rates above 5x10^{-3} Sv. In Switzerland, the Effective Equivalent Dose to the average population from natural and anthropogenic sources is about 4x10^{-3} Sv/year. In 1986, the Chernobyl fallout's contribution to the total dose rate was , on the average, 4 % or less, decreasing exponentially in future years.

Table 1. Deposition rates and deposition velocities for Chernobyl radionuclides in the Dübendorf area.

Nuclide	Halflife	Deposition rate r_D	Deposition velocities [cm/s]			
			Total Deposition		Dry Deposition	
	[Days]	[kBq/m^2] 29.4.-22.5.86	30.4.86 (12h)	30.4.-8.5.86 (9 days)	days with fog and dew 30.4.-2.5.86	without 2-12.5.86
^{137}Cs	10^4	4	3.4	0.5	0.13	0.024
^{103}Ru	40	3	2.8	0.4	0.10	0.024
^{131}I	8	20	3.0	0.4	0.21	0.080
^{132}Te (^{132}I)	3	18	-	-	-	-
^{99}Mo (^{99m}Tc)	3	5.6	-	-	-	-
^{90}Sr	10^4	0.027	(~6.5)*	(0.7)*	-	-

* *These values were calculated using ^{90}Sr/^{137}Cs activity ratios in selected air filter and precipitation samples. For further explanations, see text.*



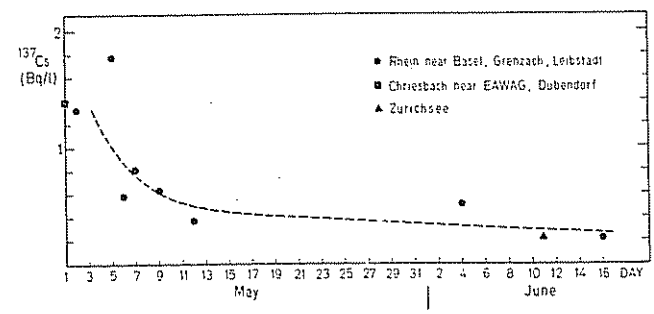
b)

4.2 Radionuclides in drinking waters and groundwaters in Switzerland

The highest concentration of ^{131}I measured in rain water was about 10^4 Bq/l and that for ^{137}Cs was about 500 Bq/l. These concentrations can be compared to maximum permissible concentrations for continuous consumption of these radionuclides in drinking water, which are 2 x 10^3 and 10^4 Bq/l, respectively. Even though the highest concentrations measured for samples of cistern water used as drinking water supply were of the same order of magnitude as for rain samples containing most of the Chernobyl fallout, their average concentration was considerably lower. However, the Chernobyl radionuclides were generally not measurable in drinking and groundwaters. One exception was reported from an experiment site near a region of infiltration of the Glatt River into groundwater [2], and where the groundwater was sampled very close (i.e., a few meters) to the river. Another exception was from the karst region of Southern Switzerland.

4.3 Washout of soils as additional input to surface waters

An important question asked is how much of the activity deposited onto catchment basins would be quickly removed via surface run-off. Even though many measurements were carried out to demonstrate that Chernobyl radionuclides were initially lost from grass at rates equivalent to 1-2 weeks residence time, very few studies exist which followed the movement of the deposited activity from soils to surface waters. None so far, however, have attempted to quantify the rate and extent of this initially rapid leaching process. In this section such a process is quantified from measurements of radionuclide activities in surface waters soon



a)

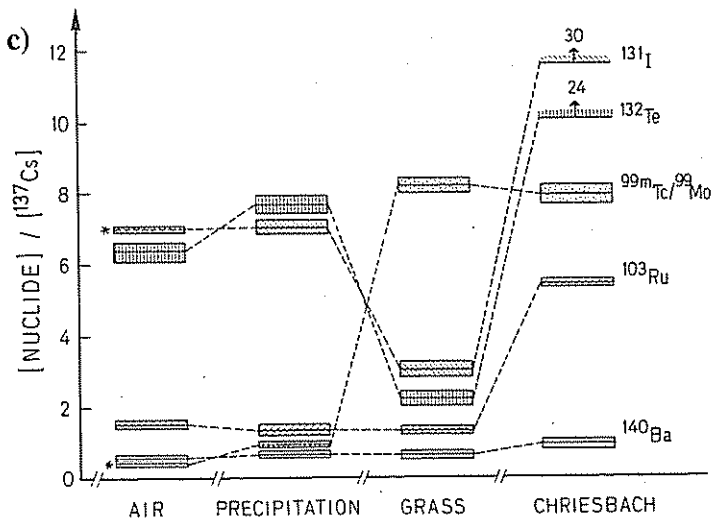


Fig. 3
a. ^{137}Cs in surface waters form northern Switzerland.
b. Nuclide to ^{137}Cs ratios in Chriesbach, next to EAWAG, and in Lake Zürich.
c. Nuclide to ^{137}Cs ratios on May 1, 1986, in Dübendorf compared for air, total precipitation, grass samples and for Chriesbach

Fig. 4
Vertical ^{137}Cs concentration profile in Lake Lugano on September 9, 1986.

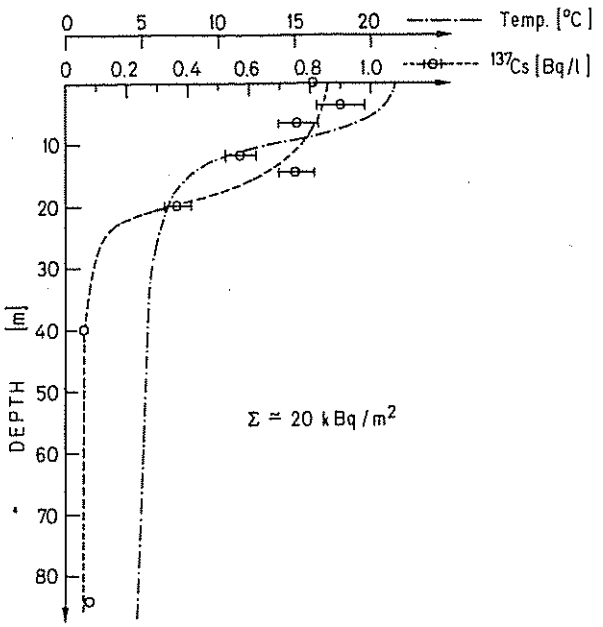
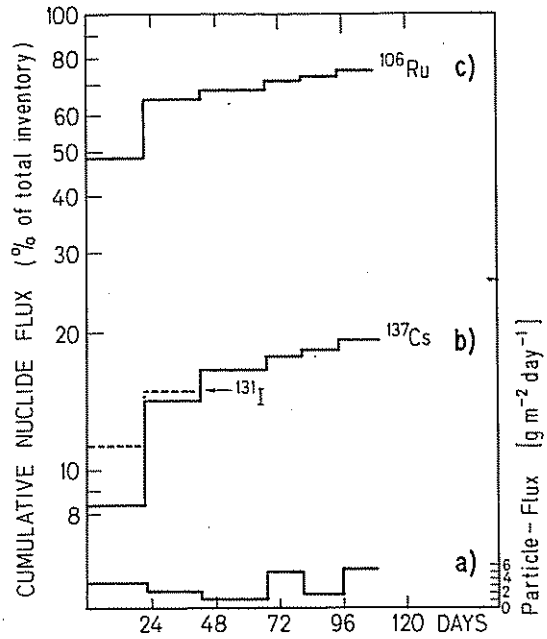


Fig. 5
Cumulative nuclide(b,c) and particle flux (a)in Lake Zürich after May 1, 1986



after deposition.

The concentrations of ^{137}Cs in various surface waters of northern Switzerland are shown in Fig. 3a, and the activity ratios of selected radionuclides to $^{137}Cs^+$, corrected for radioactive decay, are shown in Fig. 3b. $^{137}Cs^+$ concentrations in various rivers remained high during the first week after the fallout and only after they decreased to small values. Other Chernobyl radionuclides in the Chriesbach next to EAWAG showed very high concentrations only during the first few days after deposition, and quickly decreased to unmeasurably small values within a week during which ^{137}Cs activities remained approximately constant. Nuclide (^{131}I, ^{132}Te and ^{103}Ru) to ^{137}Cs ratios in the Chriesbach on May 1, 1986, were much higher (by a factor 4-5) than in rain water and air samples (Fig. 3c), presumably because these nuclides were washed out of the catchment basin as neutral or anionic species . However, $^{137}Cs^+$, a cation, also partially deposited as insoluble particles, was retained by the soil.

Due to the low adsorbability of anionic and molecular species to particle surfaces, such species can be expected to be extremely mobile in the environment. It is understandable, then, that anionic species of ^{131}I and ^{103}Ru, e.g., I^-, IO_3^-, RuO_4^{2-}, were washed off soils and vegetation, infiltrated into test wells near the Glatt River. Such species were able, also, to penetrate the surfaces of leaves and pine needles [2].

Because ^{137}Cs concentrations in surface waters were relatively constant during that time, nuclide to ^{137}Cs ratios can be taken as indicative of the relative nuclide mobility with respect to ^{137}Cs in soils.

Figures 3a and 3b show, qualitatively, that mobile anionic and neutral species of these nuclides must have existed only during the first few days after the fallout. The subsequent very low concentration of these nuclides (i.e. ^{131}I, ^{132}Te and ^{103}Ru) indicates that they must have been immobilized in the soil after a few days to a much larger degree than was ^{137}Cs. Assuming a realistic value of the surface run-off during that time of about 0.2 $cm^3\ cm^{-2}\ day^{-1}$, and typical deposition rates as described previously [2], it is p[ossible to determine that arelatively small fraction of the drainage basin of Chriesbach has a very low retention capacity, with concominant short nuclide residence times of days to weeks. Such areas correspond, in general, to terrain covered with concrete, such as streets and houses, but also rocky grounds and rivers. For ^{137}Cs, this fraction can be calculated to be on the order of 1%; and for the more mobile nuclides (^{131}I, ^{132}Te and ^{103}Ru), to be about 5-10%. These results agree, within the errors, with those by previous studies using either naturally occurring radionuclides [8] or those from bomb fallout [9]. These latter studies had determined this fraction of a catchment basin to be in the order of 1% for nuclides such as $^{137}Cs^+$ and $^{7}Be^{2+}$, which are relatively strongly sorbed onto soil particles, and about 10% for nuclides such as $^{90}Sr^{2+}$, which are sorbed onto soils to a lesser degree. A similar behavior after deposition to catchment basins can, of course, be expected for other atmospherically delivered trace contaminants. The remaining fraction of these radionuclides will be residing in the soil, however, for a very long time, i.e. about 10^3 years [8,9].

4.4 Direct deposition of Chernobyl radionuclides into surface waters

For medium-range and long-term studies, only ^{137}Cs (halflife of 30 years) and ^{103}, ^{106}Ru (halflife of 40 days and 369 days, respectively) have to be considered. Because of the long residence time of water in lakes, which is often 1 year and longer, and the relatively small contributions from drainage basins to lakes for the long-lived ^{137}Cs, we can assume that lake nuclide inventories reflect the direct atmospheric fallout to the lake area only. Since horizontal mixing in lakes is fast compared to vertical mixing, horizontal gradients should be small and one profile can be taken as representative for a whole lake. Vertical concentration profiles can therefore be indicative of mixing and elimination processes within a lake.

The ^{137}Cs inventory in Lake Zürich on June 11, 1986, calculated from a vertical concentration profile, was 4.5 kBq/m^2. This is in agreement with the atmospheric fallout measured in Dübendorf.

The ^{137}Cs profile from Lake Lugano, which is shown in Fig. 4, resulted in a

^{137}Cs inventory of 20 kBq/m^2 on September 9, 1986. Here too, this inventory agrees fairly closely with measurements of its deposition rate in southern Tessin of 21-26 kBq/m^2 [2]. In both lakes, however, ^{137}Cs had been mixed below the actual position of the thermocline at the time of measurement.

4.5 Accumulation and elimination processes in lakes

Radionuclides in lakes are taken up by particles freshly produced in the lake (plankton and $CaC0_3$) and by those washed into the lake from drainage basins (composed mostly of clay minerals). Over a time scale of months, these radionuclides are transported to the sediments by the downward conveyor belt of particles. An important route for eliminating more soluble nuclides such as $^{137}Cs^+$ is also the direct adsorption onto surface sediments in shallow parts of a lake [10]. One aim of the present studies is to differentiate between these two pathways.

Longer-lived nuclides can be accumulated by fish via food (plankton, periphyton) and water. The bioconcentration factor for ^{137}Cs in selected fish (= activity in fish/activity in water) of about $2x10^3$ cm^3 per g wet weight [2] is similar to that of plankton and is mostly gained by the $^{137}Cs^+$ taken up through ingestion of planktonby the fish [11]. This behavior of $^{137}Cs^+$ agrees with that of other radioactive trace elements released by reactors and through atomic bomb testing, which are not magnified in the food chain.

The bioconcentration factor of $^{137}Cs^+$ on sinking plankton debris of Lake Zurich was initially higher, namely 4 x 10^4cm^3 per g dry weight. This also means that the fraction of $^{137}Cs^+$ associated with suspended particles was only a few percen, at most. This fact, and the pulse-like shape of the ^{137}Cs and ^{103}Ru flux out of the lake (Fig. 5),could indicate that about 10% of ^{137}Cs probably reached the lake in insoluble aerosol particles, and is in agreement with our filtration results of rainwater. This pattern of decreasing ^{137}Cs fluxes was not produced by decreasing sedimentation rates as sedimentation rates increased during the summer months. The fraction of ^{137}Cs which was vertically transported out of the lake water to the lower-laying sediments was 10-15% of the total inventory during the first 2 months, and only about 5% during the following 2 months.

During the same time, all other nuclides disappeared from the lake water, either due to radioactive decay or due to faster removal rates. For example, seventy percent of the total inventory of ^{103}Ru was vertically transported out of the lake within the first 2 months, stressing the strong particle affinity of Ru in lakes. Such a behavior contrasts the behavior of Ru in soils and surface waters during the first few days after the fallout, and most likely points to a reductive uptake of Ru by the plankton. The speciation in biological systems is characterized by the tendency of these systems to reduce radionuclide valence after adsorption to surfaces.

Because of their ultra-low concentrations in the environment (i.e. less than 10^{-15} M), the fate and extent of accumulation in biological systems of some of these radioactive trace elements is determined by the concentration of the stable element, or of a stable homologue (e.g., K^+ for $^{137}Cs^+$, and Ca^{2+} for $^{90}Sr^{2+}$). As both K^+ and Ca^{2+} concentrations are similar in most Swiss lakes, bioaccumulation factors in different fish species from one lake and other aquatic organisms should also be similar. Indeed, ^{137}Cs concentrations in fish from different lakes in Switzerland [2] were approximately proportional to the deposition rate from the atmosphere and this determined the values of the prevalent concentrations in the lake water and sediments. No differences between herbivores and carnivores, or between fish with pelagic and littoral habitat were discernible during 1986, because of very large variability in concentrations from all species. However, the relatively uniform average concentrations of ^{137}Cs in different fish species from a single lake probably are the consequence of the uniform tagging of the food sources, i.e., suspended and sinking particles, shallow surface sediments and periphyton, with ^{137}Cs. As the residence time of ^{137}Cs in fish is about 500 days [11], it will be difficult to detect during 1987 a significant decrease of its activity concentration in fish from Lake Lugano or from other lakes.

5. SUMMARY

Observations of the temporal trend in concentrations of Chernobyl radionuclides in atmospheric terrestrial and aquatic reservoirs near Dübendorf aided in quantifying fluxes and transfer velocities from one reservoir to another. The results from these studies add to our knowledge on radionuclide movement in the environment, for which the groundwork was layed down by the earlier observations on the fate of the radionuclides released by bomb tests, earlier reactor accidents, and by purposeful tracer experiments in lakes and ocean basins. The results from our observations should also help to predict the movement of other atmospheric trace contaminants in the environment.

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AEROBIC THERMOPHILIC PRETREATMENT FOR WASTE SEWAGE SLUDGE HYGIENISATION

Geoffrey Hamer, C. Anthony Mason and Thomas Fleischmann

1. BACKGROUND

The enforcement of increasingly stringent water pollution control legislation has resulted in increased capacity for both municipal sewage and industrial wastewater treatment by various combinations of mechanical, biological and physico-chemical process technologies in most West European countries. The major by-product of both mechanical and aerobic biological treatment processes is waste sludge, a putrefactive, aqueous suspension of biodegradable, partially biodegradable and essentially non-biodegradable solids and similarly degradable dissolved and soluble matter. Waste sludge presents a serious disposal problem in Switzerland and other countries or regions that are remote from the ocean. In Switzerland, sludge disposal is generally done by spreading stabilized sludge on agricultural land.

Conventional waste sludge stabilization technology involves mesophilic anaerobic digestion, but such processes are no longer considered, by the Swiss authorities, to be entirely satisfactory for the elimination of pathogenic organisms from the waste sludge undergoing treatment. The essential requirements of the Swiss ordinance [1] are that treated sludge deposited on agricultural land must contain neither viable worm eggs nor more than 100 Enterobacteriacea per gramme of wet sludge. Thermophilic anaerobic sludge digestion processes have been proposed as obvious alternatives to conventional mesophilic anaerobic biotreatment, but the large numbers of bioreactors recently constructed in Switzerland for the latter technology cannot easily be converted to thermophilic operation. Hence, the technology that is presently being adopted in Switzerland, comprizes a pretreatment step, prior to conventional digestion, where the preferred technology for pretreatment is self-heating, thermophilic aerobic biological processing, operating either in a continuous flow mode or a semi-continuous (fill and draw) mode. Genuine thermophilic biotreatment requires process temperatures in the range 55°-70°C. Frequently, processes operating in the thermotolerant range of microbial activity, i.e. 45°-55°C, are erroneously described as thermophilic processes, but such processes should not be considered to be entirely effective in their hygienization potential for the elimination of pathogenic bacteria. In the research work described here, which was supported by Swiss National Program 7D, it is genuine thermophilic aerobic pretreatment that is considered.

Effective sludge hygienization requires either the death or irreversible inactivation of potentially pathogenic bacteria present in the waste sludge undergoing treatment, i.e., organisms present in the hygienization process feed must be destroyed by the actions of the thermophilic process bacteria responsible for mediating the sludge treatment process. Bacterial death and/or destruction, whether in technical-scale waste sludge treatment processes, during growth processes under either optimized or unbalanced conditions or as a result of externally applied stresses, has been largely ignored and is little understood when compared with the available knowledge concerning bacterial growth and growth related phenomena [2]. Much of the data concerning bacterial death is to be found in the pharmaceutical and food science literature, where contaminating bacteria present in products are subjected to extreme physical stresses such that the products are able to meet public health safety standards.

In waste sewage sludge hygienization processes growth and death/lysis are process phenomena which occur coincidently, a feature frequently overlooked and one which has led to either oversimplification of or erroneous data interpretation concerning such processes.

2. PROCESS RESEARCH

Aerobic, thermophilic hygienization processes comprize numerous unique features compared with other waste treatment processes [3]. Some of these features are microbiological in nature, whilst others are involved primarily with process engineering aspects.

The vast majority of scientific studies on both bacterial growth and biodegradation have been concerned with soluble carbon energy substrates which, given both an appropriate spectrum of other essential nutrients and sufficient time, are completely utilized to form additional bacterial biomass, carbon dioxide and, in some cases, other products, depending on whether the growth and/or biodegradation processes occur under oxygen excess/deficient aerobic or anaerobic conditions. In addition, it is also assumed that a small fraction of the carbon energy substrate is utilized for the maintenance requirements of the bacteria involved, a growth-rate independent process. In waste sludge treatment processes, the major fraction of the biodegradable matter is present as particulate solids. These are predominantly biologi-

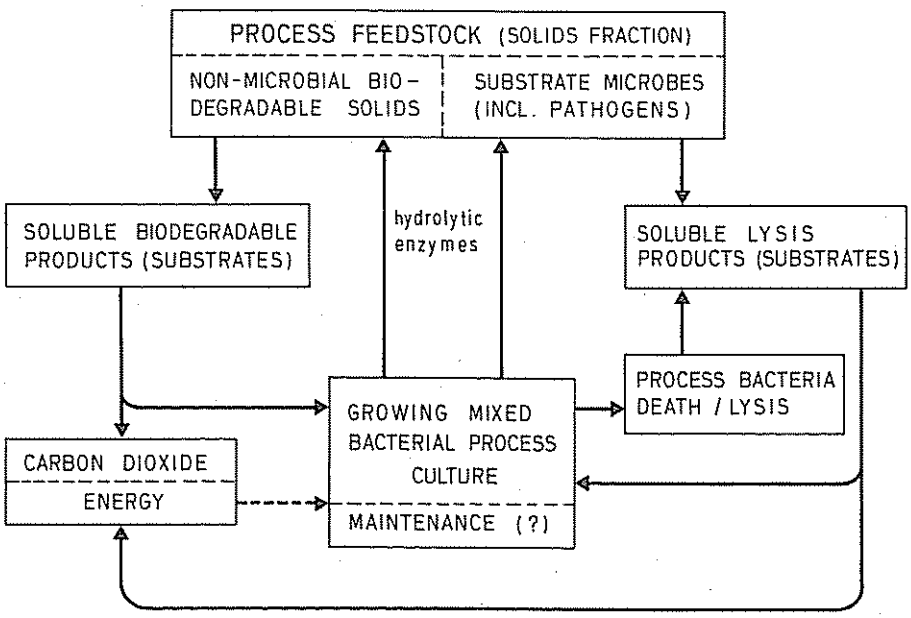

Fig. 1
Schematic diagram for the solubilization/biodegradation of the solids fraction of sludge by heterotrophic bacteria under aerobic conditions.

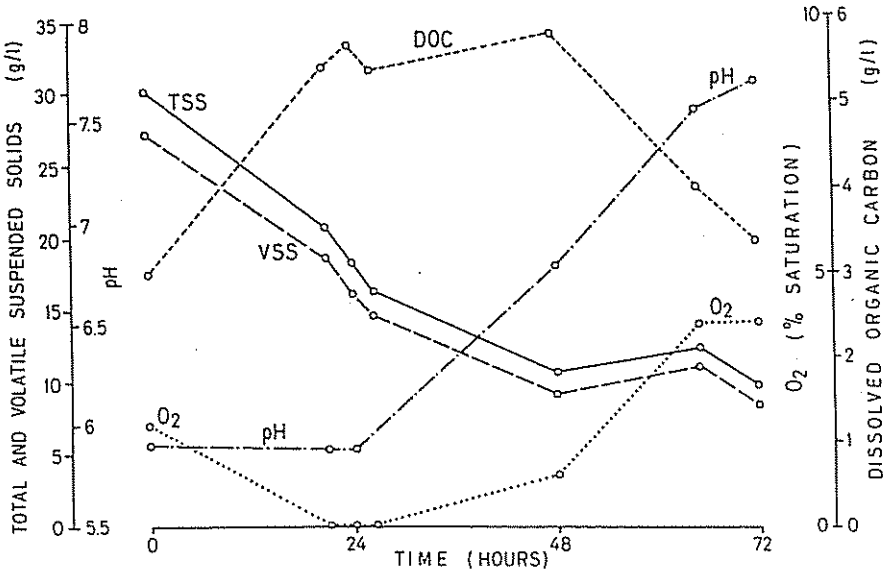

Fig. 2
Changes in the concentration of total suspended solids (TSS), volatile suspended solids (VSS), dissolved organic carbon (DOC), pH and dissolved oxygen concentration during semi-continuous aerobic thermophilic biodegradation of microbial solids with a cycle time of 3 days.

cal in origin, comprizing both microbial solids, including potentially pathogenic bacteria, on the one hand, and material such as cellulose particles, on the other hand. The utilization of biodegradable particulate matter by bacteria, as their carbon energy substrates, first involves solubilization (hydrolysis) of the solids. Clearly, the two categories of biodegradable solids mentioned are markedly different in both their structure and composition and it can be hypothesized that mechanisms involved in their solubilization will also be markedly different. In the case of cellulose particles, a modified cube root law, shrinking site type behavior can be predicted, whilst in the case of microbes, a cell wall/membrane bursting mechanism must predominate. Whilst both modes of solubilization are of importance for complete sludge treatment, it is the latter process that is required when hygienization is the primary process objective. In sludge treatment processes this involves two distinct categories of microbes, the process bacteria, on the one hand, and the substrate (feed) microbes, on the other hand. The overall process is shown diagrammatically in fig. 1.

Experimental studies have been undertaken to examine and models have been established to describe the death and lysis both of growing pure cultures of bacteria and of non-growing substrate microbes in the presence of actively growing process bacteria. In the former case, *Klebsiella pneumoniae*, a mild pathogen, was the bacterium investigated, whilst in the latter case, the only "standardized" microbes readily available, baker's yeast was used. Experiments with *K. pneumoniae* under optimized mesophilic growth conditions in continuous culture clearly indicated that in actively growing cultures, the bacteria were predominantly present as viable respiring cells, although a small fraction were present as non-viable enzymically active respiring cells. No dead, enzymatically inactive cells could be detected, and the conclusion drawn was that in such cultures, the processes of death and lysis were coincident [4]. In further studies with pure cultures of *K. pneumoniae*, the capacity of this bacterium to exhibit "cryptic" growth, i.e., growth on its own lysis products, was evaluated [5]. In these experiments, the soluble cell contents of cells taken from an optimized continuous culture of the bacterium were tested as carbon energy substrates in batch cultures inoculated with cells from the same continuous culture. Rapid growth without any lag phase was observed and it was concluded that the cells were optimally adapted to utilize their own soluble intracellular components. This finding clearly casts doubt on the concept of maintenance energy that is so widely used to explain biomass yield coefficient depression in actively growing cultures.

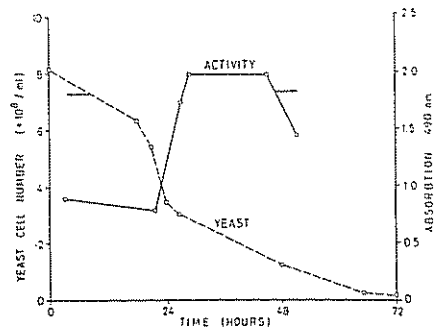

Fig. 3
Changes in aerobic thermophilic process microbe activity and substrate yeast cell number during semi-continuous aerobic thermophilic biodegradation of microbial cells.

The experiments carried out on the solubilization of yeast cells by bacterial action at thermophilic temperatures were conducted in bioreactors operating in both the semi-continuous (fill and draw) and the continuous flow modes. The mixed process culture of bacteria used was derived from a technical-scale plant operating at ca. 65°C. Its properties remained essentially stable throughout the experimental programme. Studies to determine the most effective operating conditions for microbial solids solubilization were carried out under both semi-continuous and continuous flow modes of operation. Of particular interest in the latter studies were the optimum dissolved oxygen concentration, the optimum operating temperature range and the optimum pH range that resulted in the maximum rates and levels of microbial solids solubilization. Most of the previous studies concerning dissolved oxygen suggested high dissolved oxygen concentrations, usually achieved by sparging oxygen rather than air into the bioreactor, to be the most effective for complete aerobic degradations, but in spite of this, indications were, from oxygen conversion data, that where thermophilic aerobic technical-scale pretreatment/hygienization processes were employed prior to conventional mesophilic anaerobic digestion/stabilization, using air rather that oxygen for oxygenation, oxygen restricted or limited conditions occurred in the aerobic bioreactor [6]. The laboratory studies, conducted under a wide range of dissolved oxygen concentrations, from near saturation to limited conditions, clearly indicated that the solubilization process was most effectively operated under either low dissolved oxygen concentrations or oxygen limitation [7].

Such findings are consistent with the view that effective process operation depends on the functioning not of an obligately aerobic mixed process culture, but on the combined activities of mixed process cultures comprizing both obligate aerobes and facultative anaerobes.

At low and limiting dissolved oxygen concentrations, the best operating temperature for microbial solids solubilization was 65°C, reflecting the genuine thermophilic nature of the process culture. Operation at 70°C, whilst still possible, was inferior to that at 65°C, indicating that the process culture was not caldoactive, whilst operation at 60°C, was intermediate in performance, with respect to the other operating temperatures discussed (7). The optimum pH range was between 6.5 and 7.5.

Semi-continuous operation of thermophilic aerobic pretreatment/hygienization processes is relatively widespread on a technical-scale at smaller treatment works in Switzerland and an objective of our study was to evaluate operating variables such as charge size, cycle time and the presence of an added nitrogen source on the solubilization/biodegradation of microbial solids. Operation with a charge size of 50% of the bioreactor volume resulted in the biodegradation of more than 59% of the microbial solids present in the bioreactor feed, showing enhanced performance when compared with operation where a charge size of 25% was used [8].

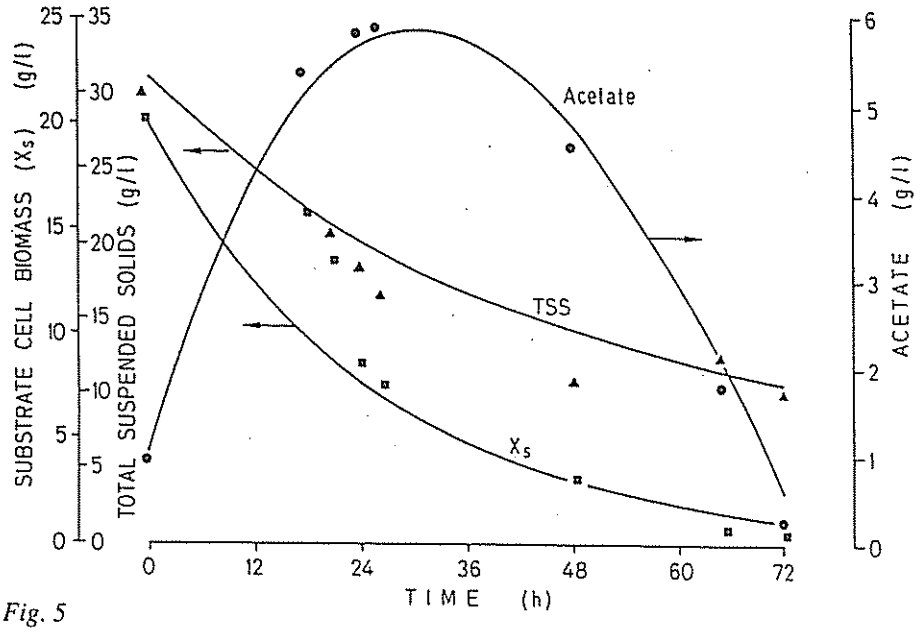

Fig. 5
Comparison of predicted values for acetate concentration, total suspended solids (TSS) and yeast cell biomass (X_s) with experimental values (o, acetate; Δ, TSS; □, yeast cells)

When microbial solids biodegradation occurs in bioreactors operating in a semi-continuous mode, the changes which occur to particular components in the system can be monitored as a function of time [9]. Some results of such analyses are shown in figs. 2-4, where a decrease in the total suspended solids concentration, shown in fig. 2, can be directly related to a decrease in the number of microbial cells used as substrate by the thermophilic process culture, shown in fig. 3. The activity of the thermophilic process culture increased during the course of the process and was probably stimulated as a result of the release of readily biodegradable substrates upon lysis of the substrate microbes. The increase in the dissolved organic carbon concentration, shown in fig. 2, occurred as a result of the production of large quantities of carboxylic acids, shown in fig. 4. Such production results from the fermentative metabolism of the thermophilic process culture, comprizing both obligate aerobes and facultative anaerobes, under oxygen restricted conditions. As can be seen from fig. 4, the carboxylic acids are sequentially degraded according to their molecular weight and structure, although both n-butyric and i-valeric acid, which are unpleasantly odoriferous, tend to accumulate and persist, a distinct process disadvantage if such processes are used without a subsequent anaerobic stage. In multi-stage processes, carboxylic acids are favored substrates that are readily biodegraded during anaerobic digestion/stabilization, thereby enhancing the rate of this stage and, hence, reducing the necessary residence time in the stage.

On the basis of the results obtained during semi-continuous operation, a mathematical model of the bio-solubilization process for microbial solids under oxygen restricted conditions, was proposed. The essential steps that were considered to occur were:

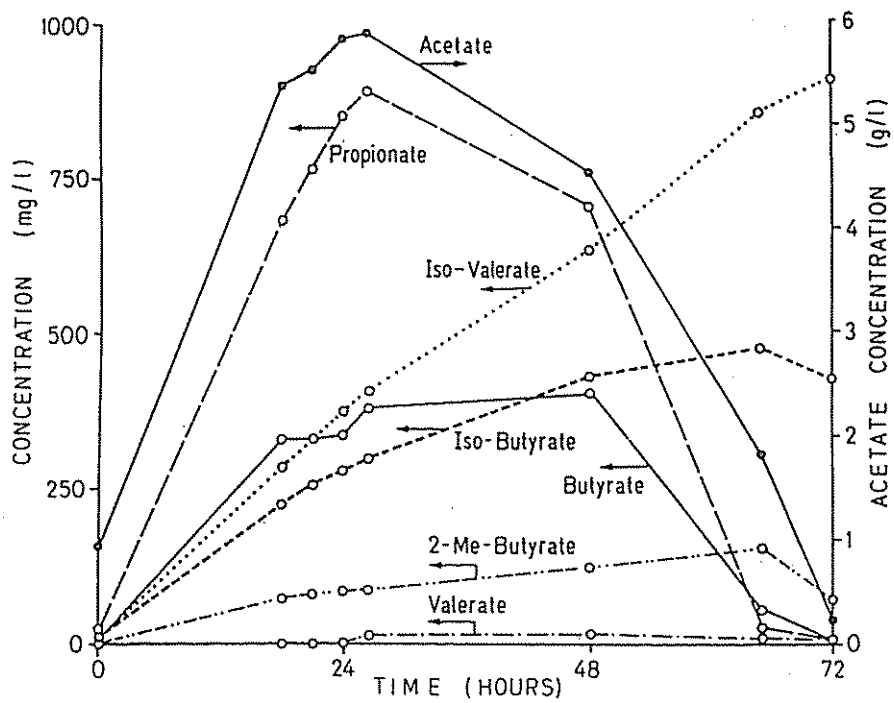

Fig. 4
Variation in the concentration of low-molecular weight carboxylic acids during the biodegradation of whole yeast cells by aerobic thermophilic microbes.

(i) A feed, comprizing a suspension of intact yeast cells, was added to the bioreactor containing an active thermophilic bacterial process culture;
(ii) The thermophilic process bacteria produce extra-cellular enzymes, which are capable of cleaving the cell walls of the yeast (substrate microbes), after non-specific attachment over the entire surface of the yeast cells;
(iii) Cell wall lysis, resulting from at least one cleavage in the cell wall, but possibly multiple cleavages before the wall is sufficiently weak to lyse, occurs and results in the release of soluble cytoplasmic components from the substrate microbes;
(iv) The thermophilic process bacteria utilize the soluble compounds as carbon energy substrates and in so doing produce carboxylic acids, predomonantly acetate, whilst further lysis of remaining yeast cells provides a continuous supply of soluble carbon energy substrates, supplemented by soluble matter resulting from the enzymic hydrolysis of yeast cell wall polymers;
(v) After significant time elapse, the rate of supply of soluble substrates from yeast cell lysis decelerates and the soluble substrate concentration falls, because production occurs only from cell wall polymers, such that the produced acetate that has accumulated is now used as carbon energy substrates by the process bacteria.
The process is considered to exhibit the molar stoichiometry of table 1, on a carbon, hydrogen and oxygen basis, with nitrogen and ash omitted.

$CH_{1.8}O_{0.43} + 1.12\ O_2$	$\rightarrow 0.3\ CH_{1.8}O_{0.43}$
Substrate Microbes	Process bacteria
	$+ 0.23\ CH_3COOH + 0.24\ CO_2 + 0.48\ H_2O$
	Acetic acid
$CH_3COOH + 2.36\ O_2$	$\rightarrow 0.38\ CH_{1.8}O_{0.43} + 1.62\ CO_2 + 3.32\ H_2O$
Acetic acid	Process bacteria

Table 1

In order to describe the growth of the thermophilic process bacteria, Monod type saturation kinetics were assumed for growth on the soluble lysis products and Monod kinetics, with an inhibition term, for growth on acetate. The remaining rate expressions, for yeast cell lysis, for process bacteria death/lysis and for residual cell wall particulate matter hydrolysis were all assumed to be first order. Incorporating appropriate rate constants and yield coefficients resulted in the predictions shown in fig. 5 for the decay in both total suspended solids and substrate microbe concentrations and in the accumulation and decay of the acetate concentration with respect to elapsed time during semi-continuous operation. Superimposed on the predicted curves in fig. 5 are appropriate experimental points, demonstrating close agreement between hypothesis and experiment and, hence, a basis for process predictions [9].

3. SLUDGE HYGIENIZATION

Most pathogenic organisms have optimum temperatures for growth below 45°C and are either inactivated or "killed" at temperatures in excess of 55°C, hence, the susceptability of pathogens to thermophilic treatment processes. Bacteria of enteric origin, especially *Escherichia coli* are used as indicators of more serious pathogens such as *Salmonella* spp. The ability of bacteria to survive heat treatment is a function of the exposure time, the temperature and the physiological status of the bacteria as a consequence of the conditions pertaining to their growth or survival during the time immediately preceding the heat shock. The realisation of effective sludge hygienization involves two major facets:
(i) The death/irreversible inactivation of pathogens;
(ii) Prevention of subsequent reinfection with pathogens and regrowth of potential pathogens.
To investigate the former, the bacterium *K. pneumoniae*, a mild pathogen commonly found in sewage, was grown in aerobic chemostat culture at its optimum temperature, 35°C, pH 6.8 and a dilution rate of 0.1 h^{-1}. The effluent from the chemostat was continuously fed into a second aerated bioreactor operating at various temperatures between 35° and 60°C, pH 6.8 and a dilution rate of 0.03l h^{-1}, in order to assess survival potential during continuous flow operation without any additional nutrient feed.
The effect of the operating temperature in the second bioreactor on the steady state bacterial dry weight, bacterial cell number, irrespective of their physiological status, and the metabolic activity are shown in fig. 6. Compared with the growing culture, the change in metabolic activity was most pronounced even at 35°C. However, what is most important to note is that at 55°C and even at 60°C and mean residence times in the second bioreactor of 32 h, the metabolic activity, although very low, was not in fact zero. Obviously, this results from the continuous feed of viable cells to the bioreactor, but the implication is that in such continuous flow completely mixed systems, it would be possible for pathogens to bypass inactivation, suggesting that such

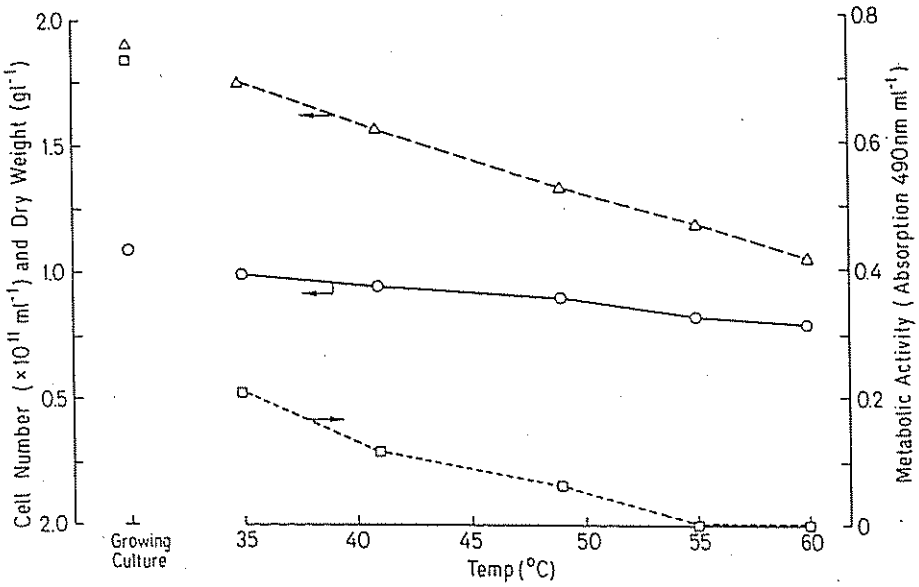

Fig. 6
Changes in dry weight (o), metabolic activity (□) and cell number (Δ), as a result of heat treating a culture of K. pneumoniae at various temperatures.

a mode of operation does have its limitations as far as complete hygienization is concerned [10].

In order to evaluate the question of regrowth potential, operation as described above was repeated at 49°, 55° and 60°C. During steady state operation at each temperature, the flow of effluent from the chemostat into the second bioreactor was interrupted and the second aerated bioreactor was maintained at its operating temperature for 4 hours. The temperature of the second bioreactor was then reduced to 35°C and fresh medium was continuously fed to the bioreactor, such that the dilution rate was 0.08 h^{-1} After an initial period, where wash-out occurred, it can be clearly seen in fig. 7 that, for each operating temperature employed in the second bioreactor during two-stage operation, regrowth of *K. pneumoniae* occurred.

The most likely explanation for this is that a potential source of reinfection existed in the head-space of the incompletely filled bioreactor used, as a result of aerosol carriage of bacteria from the aerated liquid to the walls of the head space where accumulation occurred. The temperature of the walls of the head space is significantly lower than that in the bulk liquid and, hence, survival is possible. In technical-scale thermophilic aerobic hygienization processes designers will have to face the problem of pathogen transfer to cool regions of the bioreactor as a result of foaming by either effective foam breaking coupled with head-space wall heating or operation under completely fitted conditions.

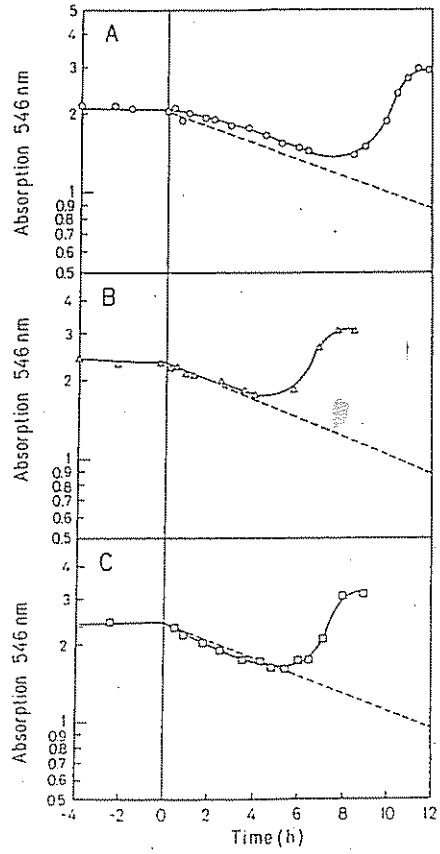

Fig. 7
Regrowth of heat treated cultures of K. pneumoniae. Continuous feed of microbes was halted four hours before returning the temperaure to 35°C and starting nutrient flow. A: 49°C; B: 55° C; C: 60°C. The expected wash-out curves are also shown (- - -)

4. CONCLUDING REMARKS

Thermophilic aerobic pretreatment processes provide one technology for achieving effective hygienization of waste sewage sludges. However, both process design and operating strategies must be based on an adequate understanding of the process microbiology of such systems. Moreover, aerobic thermophilic hygienization processes must be coupled with fully effective stabilization processes if treated sludge is to meet stringent standards with respect to pathogens elimination.

It is unlikely that the use of aerobic thermophilic processes for complete sewage sludge stabilization, as independent processes, will compete with conventional anaerobic mesophilic treatment on either an economic or a technological performance basis, although their use as a pretreatment process could, in addition to achieving hygienization, also increase process rates during subsequent anaerobic digestion/-stabilization.

Our continuing research programme on aerobic thermophilic pre-treatment for waste sewage sludges is concentrated on two further objectives:

(i) The coupling of an optimized pretreatment process with an optimized mesophilic anaerobic digestion/stabilization process;

(ii) Minimization of the biomass yield coefficient for aerobic thermophilic cultures of the process bacteria, thereby reducing the load of biodegradable solids transferred to anaerobic digestion/stabilization.

Geoffrey Hamer is Professor of Technical Biology at the Swiss Federal Institute of Technology Zurich (ETHZ) and Head of the Technical Biology Department at the Swiss Federal Institute for Water Resources and Water Pollution Control (EAWAG). His major research interests are in applied microbial physiology, environmental biotechnology and bioprocess engineering.

C. Anthony Mason recently obtained his doctorate at ETHZ for research undertaken in Prof. Hamer's laboratory on the subject of this article. He is presently a post-doctoral research fellow in the Chemical Engineering Department of the California Institute of Technology in Pasadena.

Thomas Fleischmann is a research technician in the Technical Biology Department of EAWAG.

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CONTRIBUTION OF BACTERIA TO RELEASE AND FIXATION OF PHOSPHORUS IN LAKE SEDIMENTS

René Gächter and Antonin Mares

It is generally believed that lake sediments act as a sink for phosphorus (P) under aerobic conditions and that large amounts of P are released when sediments become anoxic. For over 40 years it has been assumed that this cycling is mainly controlled by inorganic processes as described by Einsele [1] and Mortimer [2], where orthophosphate combines with oxidized iron to a solid phase and later becomes released if Fe^{3+} is reduced and redisolved. According to this traditional model sediment microorganisms only play an indirect role by lowering the redox potential while decomposing organic matter and thus providing the necessary condition for reduction of iron and subsequent release of phosphate.

Although numerous results from field and laboratory studies are in convincing agreement with this model, there are some results reported in the literature [e.g. 3-5] which cannot be explained by it solely.

Most studies ignore that bacteria grown under aerobic conditions are able to store large amounts of phosphorus and then are able to release it very quickly if their environment becomes anoxic [6,7].

In the experiment described in fig. 1 sediment bacteria were grown in an aerated medium containing 6.7 g standard nutrient broth (Merck), 1 g glucose and 0.27 g NH_4Cl per liter of filtered and sterilized lake water. When the dissolved phosphate was exhausted, the culture was subdivided into two parts. To one part, particulate iron in the form of freshly precipitated iron-oxihydroxide was added. Then, in order to remove the oxygen, both cultures were bubbled with nitrogen. As fig.1 demonstrates, iron and phosphate dissolved simultaneously although - for experimental reasons- iron and phosphate never were associated together in the solid phase before the dissolution.

After bubbling with air, the released SRP was immediately reconverted into particulate phosphorus in both cultures. Based on these results we propose an antithesis to the purely abiotic model mentioned, suggesting that

- the phosphorus cycling at the sediment/water-interface is controlled to a large degree by microorganisms and
- the geochemical release of iron and the biological release of phosphate are not coupled, but appear to happen simultaneously.

Preliminary results obtained from sediments of Lake Sempach [8] indicate that up to 40% of the total phosphorus is incorporated in microbial biomass. Thus, the release of SRP from lake sediments cannot simply be described by inorganic pH- and redox-potential dependent solubility equilibria and by diffusion. Production and decomposition of microbial biomass and biological uptake and release of soluble reactive phosphorus (SRP) by bacteria seem to be other important processes which need to be considered.

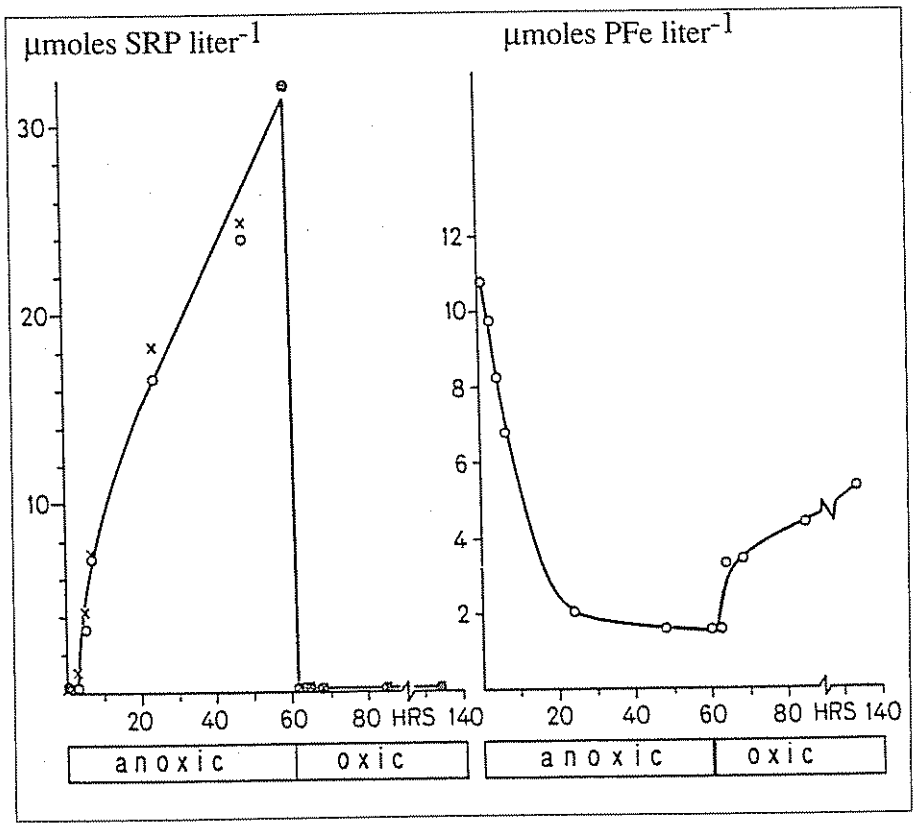

Fig. 1
Uncoupled but simultaneous release of phosphorus and iron from an anoxic culture of bacteria
Left: Mobilisation and immobilisation of soluble reactive phosphorus (SRP) are independent of whether iron oxihydroxide flocs were added (o) or not (x).
Right: Dissolution and formation of particulate iron (PFe).

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ORGANIC MICROPOLLUTANTS IN THE ATMOSPHERE:
Determination, Origins and Behavior in Rain, Snow and Fog.

Walter Giger, Christian Leuenberger, Jean Czuczwa and Josef Tremp

1. Organic Trace Constituents in the Atmosphere

Besides the major components nitrogen and oxygen, the atmosphere contains a complex mixture of inorganic and organic constituents including water, carbon dioxide, noble gases and ozone as well as methane and other organic compounds. These atmospheric trace components play an important role in many terrestrial processes, some of the most important examples of which are weather, radiation balance, photosynthesis and protection from solar radiation.

Organic compounds enter the atmosphere from various emission sources, from which both biogenic substances (methane, terpenes) and above all pollutants caused by human activity are of interest (fig. 1). Emissions from automobiles, household heating, industrial sources, municipal incineration and agricultural activities contribute organic contaminants to air pollution.

In the atmosphere the primary pollutants are subject to complex dilution, transport, dispersion and transformation processes (transmission) and can be converted into secondary pollutants through photochemical reactions. The atmospheric concentrations which finally result and the effects on ecosystems including man (immissions) are characteristically influenced by chemodynamic restraints on the pollutants. Of particular significance is the physical state of the pollutant entering the atmosphere. The behavior during transmission is much different for a gaseous compound in comparison to a particle-bound substance.

Condensed atmospheric water (rain, fog and snow) can remove pollutants by gas scavenging as well as by particle scavenging. The distribution processes between gaseous, dissolved and particulate phases are important in determining the behavior of atmospheric pollutants. These processes especially influence the extent of damage from atmospheric pollution to terrestrial and aquatic ecosystems through wet and dry deposition.

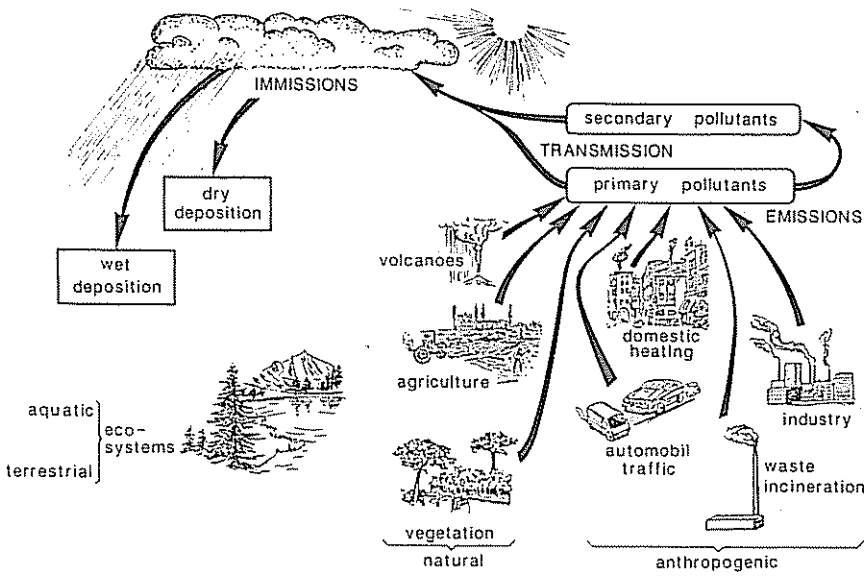


Fig. 1
Organic air pollutants: from emission to immission.

2. Environmental Pollutants in Atmospheric Precipitations

Within the framework of the National Research Program 14, "Atmospheric Processes and Air Pollution in Switzerland", a research project is being carried out at EAWAG. The primary goals of this project were the development of dependable procedures for sampling of wet depositions and for quantitative determination of selected classes of organic substances. With the help of these methods the concentrations of organic compounds in wet deposition (rain, snow) and in fog were measured. The separate determination of dissolved and particulate fractions was especially emphasized.

A specially developed instrument was available for rain collection (Pankow sampler). This accomplished simultaneous filtration for separation of particles as well as enrichment of rain-dissolved substances on an adsorption column. Chromatographic and spectroscopic methods were employed for the determination of individual organic

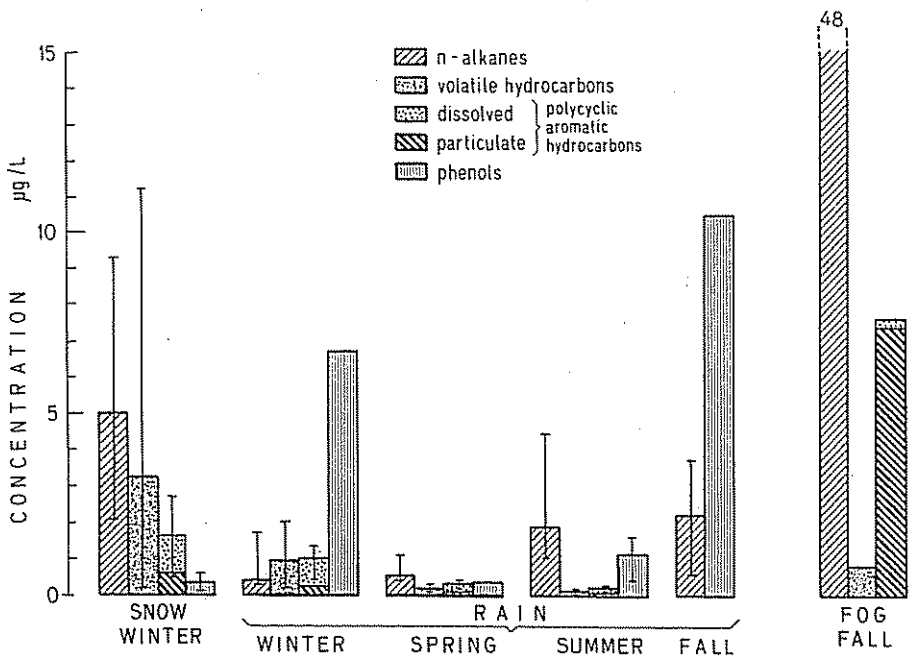


Fig. 2
Average and distribution of concentrations of classes of organic compounds in rain, snow and fog. The concentrations refer to water volumes.

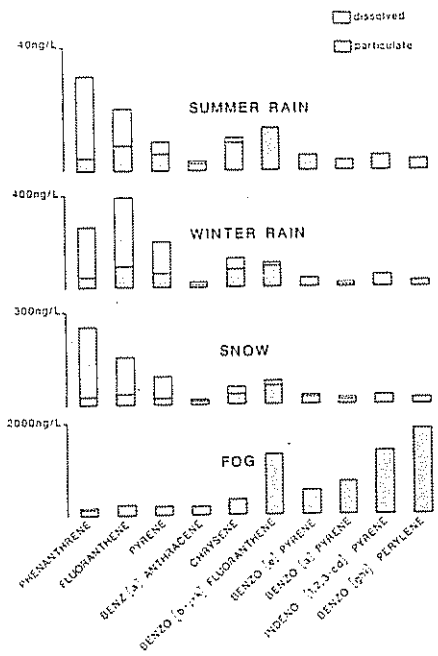


Fig. 3
Concentrations of individual polycyclic aromatic hydrocarbons in summer and winter rain, snow and fog. The concentrations refer to water volumes.

compounds (GC, GC/MS, HPLC). Figures 2 through 6 contain an overview of the most important results of this research. The loading of precipitation with alkanes, volatile hydrocarbons, filterable and particulate phenols and polycyclic aromatic hydrocarbons (PAH) each have their own distinct characteristics with respect to precipitation type and season of sample collection (fig. 2). The different sources of individual classes of compounds lead to different modes of emission and dissimilar atmospheric behavior. The relatively high phenol concentrations in the rain samples indicate effective scavenging of the well soluble phenols. The extremely high loadings of fog water with alkanes and PAH are especially noteworthy. However, since only two fog samples have been analyzed, these values must not yet be considered highly certain.

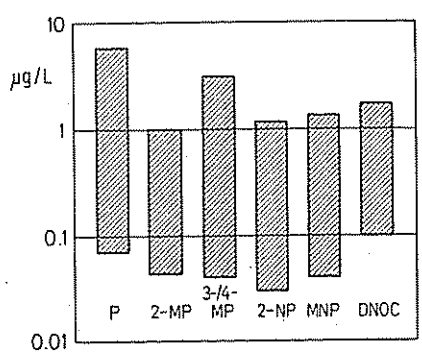


Fig. 5
Distribution of concentrations of phenols in rain and snow. P: Phenol; MP: methylphenol; NP: nitrophenol; MNP: methylnitrophenol; DNOC: 2,4-dinitro-*ortho*-cresol.

The gas chromatographic analysis of saturated hydrocarbons (alkanes) yields clear information about the origin of these substances. All filtered material (>0.2 µm particle diameter) contained the homologous series of straight-chain saturated hydrocarbons (*n*-alkanes) with chain lengths ranging from 15 to 35 carbon atoms (n-$C_{15}H_{32}$ to n-$C_{35}H_{72}$). N-Alkanes of biogenic origin dominated in the particulate alkanes from the summer samples. In winter rain and in fog, however, alkanes originating from oil or combustion products were predominant. It should be noted that saturated hydrocarbons should be found exclusively associated with particles because of their poor water solubility. The presence of colloidal adducts could explain their elevated occurrence in the dissolved fraction in rain.

Trace concentrations of alkylbenzenes (toluene, ethylbenzene, xylenes) and volatile chlorinated hydrocarbons (tetra- and trichloroethylene, 1,4-dichlorobenzene) were also observed. The alkylbenzenes belong to the fraction of gasoline and of fuel oil which is water soluble. Finally, considerable amounts of the chlorinated solvents trichloro- and tetrachloroethylene (used for dry cleaning and metal degreasing) is lost to the atmosphere during normal application.

About thirty PAH were determined qualitatively and quantitatively. The PAH concentrations increased with precipitation type as follows: summer rain < winter rain ≈ snow << fog water (fig. 3). In the rain and snow samples the three and four ring PAH dominated; in fog, PAH with five and six rings were the main components.

The proportion of PAH associated with particles increased with decreasing size of the PAH molecule. The partition coefficient D between the dissolved and particulate form in rain samples yielded a correlation with the octanol-water coefficient (fig. 4), indicating a relationship between the dissolved-particulate phase partitioning and the solubility for individual PAH. This correlation results from the interaction of various physicochemical processes responsible for phase distribution. Differences in gas and particle

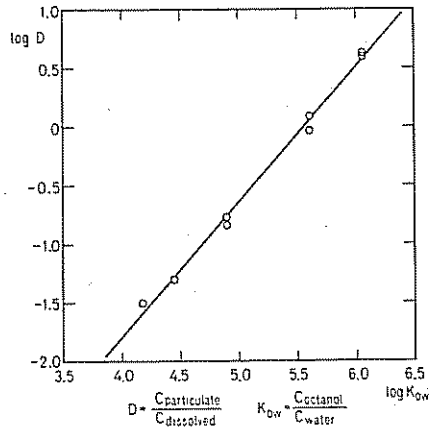


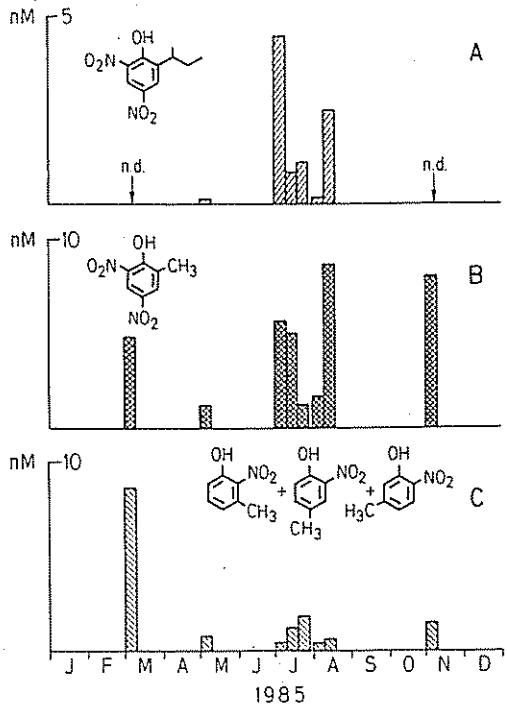
Fig. 4
Correlation between the ratio of particulate to dissolved polycyclic aromatic hydrocarbons in rain and their octanol-water coefficients (K_{OW}).

scavenging processes between individual PAH are especially significant. Hydro- and lipophilicity of a pollutant are also important factors in determining the phase distribution.

Rain contained phenol and methylphenols (cresols) in concentrations ranging from 0.022 to 3.8 µg/l (fig. 5). In addition, nitro- and dinitrophenols were also found in concentrations approaching these levels. All phenols occurred in the filterable part of the precipitation. The phenols and cresols are due to engine combustion emissions with a possible contribution from secondary formation during atmospheric transmission. The origin of the nitrated phenols is not yet fully explained. Certain dinitrophenols (DNOC, Dinoseb) could originate from agricultural activities since they are used as pesticides. Dinoseb exhibits elevated concentrations in summer rain. In contrast, DNOC shows no seasonal differences, as would be expected for agricultural emissions (fig. 6). Other observations suggest that the nitro- and dinitrophenols result from automobile emissions, since formation from aromatic hydrocarbon precursors in the presence of hydroxyl radical ions and nitrogen oxides is possible.

Until recently the importance of all organic air pollutants was primarily judged with reference to their harmful effects on man. For this reason cancer causing PAH such as benzo[a]pyrene stood at the forefront. Further attention has been given to the problem of a possible decline of the stratospheric ozone layer through the extremely high persistence of the

Fig. 6
Seasonal fluctuations of concentrations of nitrated phenols in rain.
A: dinoseb; B: DNOC; C: methyl-2-nitrophenols



chlorofluoromethanes (Freons). Motivated by the forest decline in central Europe, which cannot be explained by a single cause, organic air pollutants with possible ecotoxicological effects (e.g., peroxyacetyl nitrate and other photochemical oxidants) are now also evaluated In this context, the results concerning trace concentrations of nitrated phenols should be discussed. Nitrated phenols and above all dinitrophenols are biologically very active molecules which uncouple oxidative phosphorylation and disturb cell metabolism already at low concentrations (1 µM - 1 mM). Such toxic organic substances in atmospheric deposition could represent stress factors for the terrestrial and aquatic ecosystems which have until now not been recognized.

3. Consequences for the Protection of the Environment

The research results will be evaluated with regard to their significance for practical environmental protection measures In Switzerland organic air pollution is addressed through clean air regulations and emission controls. The imminent revision of the ordinances for emission controls is expected to have a greater probability than before of decreasing the concentration of organic pollutants occurring in atmospheric precipitations.

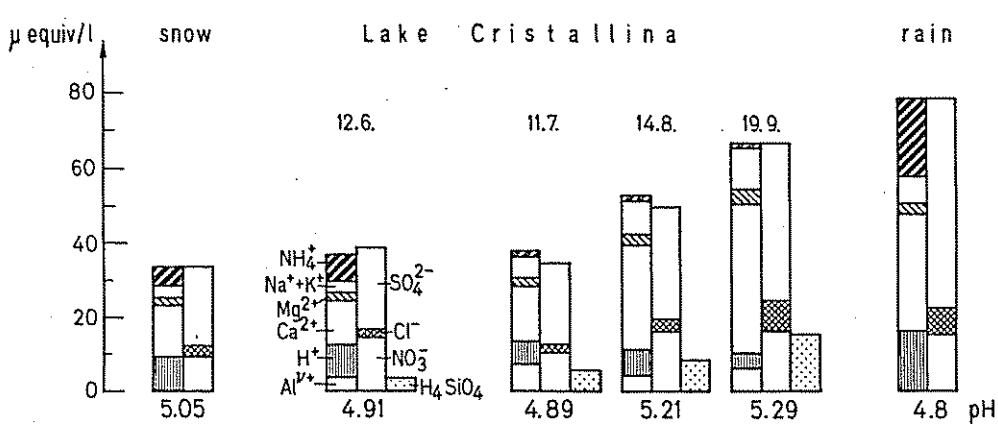
BUFFERING MECHANISMS IN ALPINE LAKES

Jürg Zobrist, Laura Sigg, Werner Stumm and Jerald Schnoor (University of Iowa)

Small alpine lakes which are situated above the timberline, where there is little soil, represent a simple natural system for studying the interaction of acid atmospheric deposition with crystalline bedrocks such as gneiss and granite.

Generally, the acidity measured in surface waters results from the difference of processes occurring in the drainage bassin, i.e. of those that produce and of those that consume protons. In high alpine areas the protons produced originate mainly from wet and dry atmospheric deposition and from transformation reactions of ammonium (nitrification and assimilation) which is also supplied from atmospheric deposition, whereas chemical weathering of rocks represents the dominant process that uses up protons. This proton balance can be expressed quantitatively assuming known elementary reactions for the weathering of silicate minerals. For example, in the drainage area of Lake Cristallina the proton input by the atmospheric deposition amounts to 17 mequiv/m^2y, while the contribution from ammonium assimilation is 12 mequiv/m^2y. This yields a total of 29 mequiv/m^2y. By comparison, the weathering of crystalline rocks neutralizes only 24 mequiv/m^2y of protons. Therefore, an excess of hydrogen ions remains in the catchment basin and the result is an acidification of the lake and its river waters.

Conclusion: In alpine crystalline areas, the low weathering rate of silicate rocks is insufficient to neutralize any more the increased atmospheric deposition of strong acids and acid-producing substances.



Seasonal variation in the water composition of the high alpine Lake Cristallina (surface area 0.75 ha, drainage area 17 ha) during summer 1985 in comparison with the estimated atmospheric deposition (derived from precipitation data mea- sured 10 km WSW).
In the lakewater, the increase of the concentrations of the cations Ca^{2+}, Mg^{2+}, Na^+, K^+ and of H_4SiO_4 is due to the interaction of crystalline rocks with protons (using up protons, means restoring the pH during the summer to the expected value of about pH 5.4). The soluble Al(III) concentration is controlled by the dissolution equilibria of gibbsite. The concentrations of the anions are regulated by atmospheric deposition.

SIMULATION OF COMPLEX ACTIVATED SLUDGE PLANT FLOW SCHEME

W. Gujer

The discharge and the operating requirements of biological waste water treatment become more and more complex. Organic carbon, nitrogen and phosphorus compounds must reliably be removed by a variety of biological, chemical and physical processes. For activated sludge systems this requires - that already in the phase of plant design- it is necessary to consider different flowschemes and load variations. Mathematical simulation with the aid of computer programs is an ideal tool for this task. Further, such programs may be used to analyze and improve the operation of existing plants.

On the basis of the bio-kinetic model for complex activated sludge systems as proposed by the IAWPRC (International Association for Water Pollution Research and Control) Task Group for Mathematical Modeling for Design and Operation of Biological Waste Water Treatment a simulation program was developed which may run on IBM compatible personal computers and which may, with small effort by the user and with good accuracy, predict the performance of different flow schemes under a variety of operating strategies under diurnal as well as weekly or even seasonal flow and load variations.

At the moment this model/program simulates organic carbon degradation, nitrification, denitrification, sludge production, oxygen consumption and soluble pollutant concentrations along a cascade of CSTR type aeration tanks and in a secondary clarifier. The model/program has been verified for a wide range of applications with experimental results from domestic waste water treatment. In further projects:

- degradation of NTA is also included in the model/program with good results.
- a variety of control loops may be defined and their performance in plant operation may be tested.
- further process flow schemes, such as two step activated sludge plants, discontinuous processes, aerobic sludge stabilization etc. are built into the program and further expand its potential.

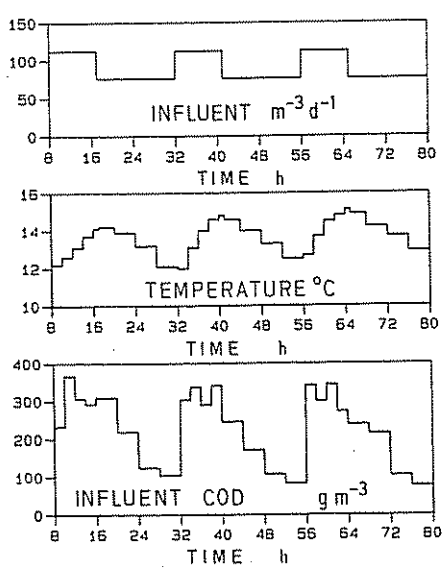
An example of the verification of the model with regard to diurnal variation of effluent ammonia concentrations from a nitrifying single CSTR type pilot scale activated sludge process is given in fig. 1. The model parameters for this simulation were obtained from experiments with domestic waste water as published in the international literature [1], and no calibration was necessary to obtain the results of fig. 1.

A sample of a potential application of this program is demonstrated in figure 2 [2]. For an activated sludge system with three equal reactors in series, the ammonium concentration and the total oxygen consumption are illustrated for a period of four days subject to significant load variations. It is obvious, that this type of information may help to improve the design of biological waste water treatment plants.

A further application is in operation of existing plants. In the context of consulting work, the program is used today in order to test the reliability of existing performance data. Frequently simulation of the operation of existing plants helps to identify systematic errors in data collection , or how the existing facilities may be used in order to obtain improved treatment performance. For instance, oxygen limitations during high load periods which may appear in some parts of the aeration tank can easily be identified. Improved distribution of aeration capacity or feed flow may be analyzed to improve the situation.

The application of the simulation program in practice (consulting firms) has been tested with several specifically chosen and trained engineers with good success. Given enough time, to understand the details of the bio-kinetic model, engineers involved in process design can soon make beneficial use of the program and are enthusiastic on the potential of this new tool. For many practical engineers, this is the first time, that they fully realize the dynamic nature of biological waste water treatment. Simulation provides them with a tool which allows to gain significant information from existing facilities, which can then be used to improve the design of upgraded treatment plants.

In education of graduate students, simulation programs allow for efficient transfer of information. In short simulation sessions, the students are exposed to the effects and performance of a vast variety of operating conditions and flow schemes of the



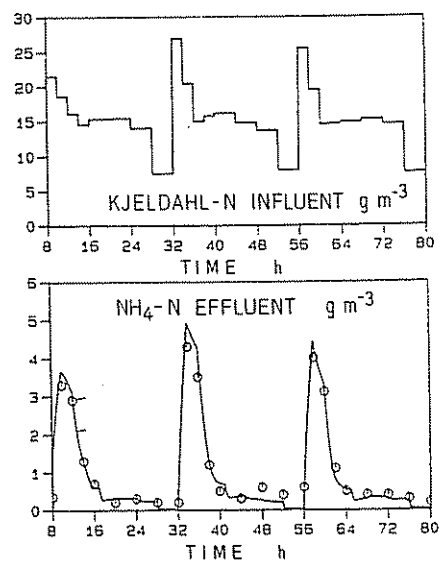


Fig. 1
Comparison of simulated (continuous line) and observed (data points) ammonium concentrations in the effluent of a completely mixed , pilot scale , activated sludge reactor operated at an SRT of 5.4 days. The diurnal variation of the influent conditions for flowrate, total COD and Kjeldahl Nitrogen as well as the reactor temperature served as controling parameters. Original data by EAWAG (1975).

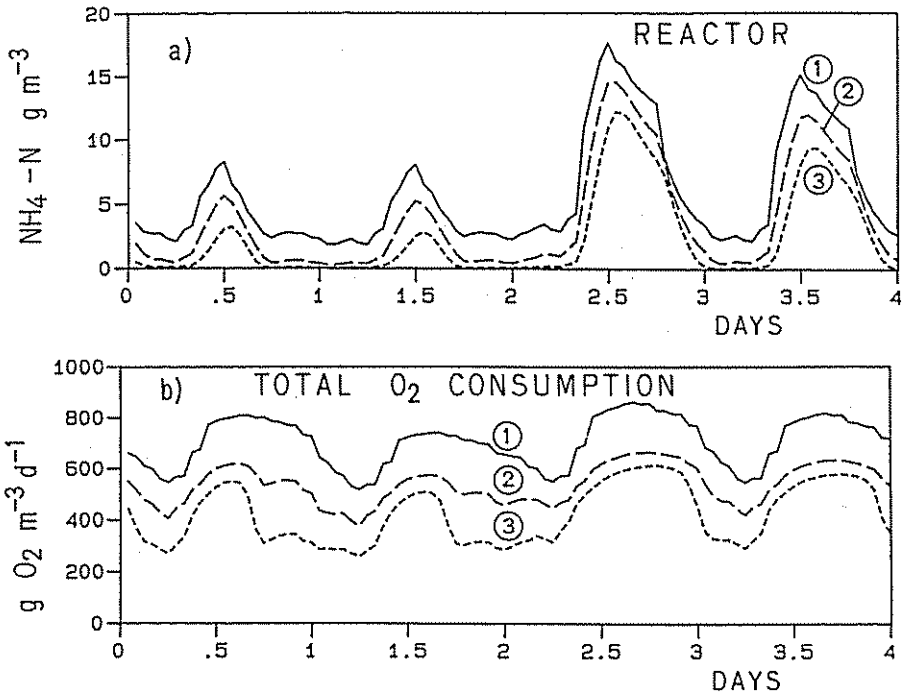


Fig. 2
Simulated results for an activated sludge process with three equal completely mixed reactors in series, subject to variable loading conditions over four days.
a) Variation of the ammonium concentrations in the three reactors 1, 2, 3.
b) Variation of the total oxygen consumption in the three reactors (Details are given by Gujer 1987).

activated sludge process. Simulation can of course not substitute practical experience, but it can enhance the conceptual understanding especially with regard to the dynamic behavior of the process. Simulation programs are used with good success in an application oriented problem solving course taught at EAWAG in the context of the graduate program in sanitary engineering and water pollution control of the Swiss Federal Institute of Technology in Zurich.
In cooperation with a private consulting firm, the program will now be brought into a form which allows for commercial distribution. An english version of the program will become available in fall 1987. In courses lasting three days and organized by EAWAG users will be introduced to the program.

[1] Gujer, Willi (1985) "Ein dynamisches Modell für die Simulation von komplexen Belebtschlammverfahren" Habilitation Thesis, Swiss Federal Institute of Technology, Zurich.
[2] Gujer, Willi (1987) "Die mathematische Simulation von Belebungsanlagen als Werkzeug für die verfahrenstechnische Gestaltung" Schriftenreihe WAR 31, Institut für Wasserversorgung, Abwasserbeseitigung und Raumplanung der technischen Hochschule Darmstadt.

ORGANIC CARBON IN THE RESIDUES OF WASTE INCINERATION

Paul H. Brunner, Hermann Moench and Steve McDow

The incineration of municipal solid waste (MSW) produces residues (bottom ash, filter dust and scrubber sludges, flue gas), which have to be disposed of in the environment. In order to enable the safe disposal of these residues, their composition and behavior in the environment have to be known. In this paper, results are presented from an investigation of the content of Total Organic Carbon (TOC) in the products of incineration. Conclusions are drawn regarding the disposal of bottom ash and filter dust, and it is suggested to improve the mineralization degree of MSW incineration.
All products of incineration contain organic carbon. Starting with the work of Hutzinger et al in 1977 [1] it became well known that hazardous organic substances such as chlorinated dioxins and furans are present in the flue gas and filter dust of MSW incinerators. Today, the occurrence and formation of certain organic trace substances in the products of MSW incineration is better known than the amount and the composition of the much more abundant organic matrix compounds. Since this matrix can determine the behavior and fate of the trace organic substances as well as of some inorganic compounds of the incineration residue, and because the emission and/or leaching of the organic matrix itself can pose a threat to the environment, the amount and nature of this matrix should be known.
As a first step, Total Carbon (TC) in bottom ash, filter dust and flue gas of MSW incinerators was measured by methods which are described in [2,3]. From these measurements, the flux of carbon through incineration was calculated. In addition, TOC in the residues of incineration was determined as the difference between TC and Carbonate Carbon (CC). In order to determine further the nature of TOC in the ash and dust, samples were analyzed by differential thermogravimetry as well as extracted with dichloromethane. Both methods indicated that TOC as determined in this work contains large fractions of non-graphitic and non-amorphous carbon.
The results of the survey of four well operated Swiss MSW incinerators are summarized in table 1. The highest concentrations of organic carbon were found in the filter dust.The TOC concentrations in the slag were slightly lower. In order to evaluate the quantitative importance of the three sources, the specific load of TOC per mass unit of MSW incinerated was calculated from the incineration mass balance (table 1). The highest load of TOC is contained in the bottom ash, and the lowest in the flue gas. The total amount of organic carbon derived from MSW incineration (2-4 g/kg MSW) is high when compared to other com-

bustion processes. A well operated waste incinerator oxidizes approximately 99% of the carbon contained in the MSW. In a coal furnace, 99.9% of C are converted to CO_2, and in a fuel oil incinerator 99.99%.

Filter dust and bottom ash are used for road construction or are disposed of in sanitary landfills. In such deposits leachates with DOC concentrations of 10-50 mg/l have been observed. The bulk of the organic compounds in these leachates has not been analyzed yet. The biodegradable fraction of the organic carbon in the ash (unburned lignocellulosic material) is utilized by microorganisms to produce methane and carbon dioxide. Thus, the pH-value of an ash deposit may be lowered by the reaction of carbon dioxide with the metal hydroxides of the ash, favoring the mobilization and leaching of heavy metals from the ash deposits. The decrease of the pH-value with time as observed in actual ash deposits is presented in fig. 1. If the degree of combustion of the ash is improved by one order of magnitude, (i.e. TOC= 1g/kg, combustion efficiency 99.9%), biochemical reactions in the not yet fully understood reactor "ash deposit" will become of minor importance.

Values for TOC in filter dust samples are given in table 2. In contrast to the results for TOC in bottom ash, there are large differences between the values of the four incinerators investigated. This suggests that incinerator design and operation (furnace temperature, residence time, gas cooling and treatment, etc.) can have a distinct effect on the TOC in the raw offgas of a furnace, and that the mineralization during combustion can be improved by existing technology.

The fate of filter dust borne TOC in disposal sites is not yet known. Trace organic substances such as chlorinated dioxins and furans have been found in filter dust as well as leachates from ash deposits [4]. In order to assess the risk of the leaching of organic substances from dust deposits, it is necessary to know the composition of the TOC. Available information does not account for more than 0.5 % of the TOC in filter dust. These substances comprise chlorinated benzenes, chlorinated phenols, chlorobrominated benzenes and polycyclic aromatic hydrocarbons. The compounds which are most likely to be leached from a deposit are chlorinated phenols, because their pK-values range around the pH-value of an aqueous suspension of the filter dust.

In order to examine the TOC in the flue gas, a new sampling train was developed which allows the differentiation between particulate, condensable and gaseous TOC. On the particle filter, only about 10% of the TOC of approx. 40 mg/Nm^3 (11% O_2) was found, which points out that TOC in the flue gas from MSW incinerators cannot be further eliminated by particulate filtration. By condensation at -10°C and sorption on florisil approx. 40 % of the TOC were trapped. The remaining 40 - 50% proved to be highly volatile C_1 to C_6 aliphatic hydrocarbons. The large variations between the four sampling campaigns are possibly caused by the non-stationary combustion due to the rapidly changing fuel quality of MSW.

It is concluded that the mineralization of MSW should be optimized. As a new goal, 99.9% of the carbon entering a furnace should be oxidized. This may be achieved by improving the degree of incineration of the bottom ash, by increasing the combustion efficiency of the offgases, and by an additional secondary combustion of the filter dust.

Table 1
Range of TOC in bottom ash, dust from electrostatic precipitators and fabric filters, and flue gas as measured in four Swiss MSW incinerators.
1) assuming 250 kg ash per kg MSW incinerated 2) assuming 2.5 kg filter dust per kg MSW 3) assuming 6 Nm^3 flue gas per kg MSW

		bottom ash	filter dust	flue gas (11% O_2)
TOC concentration	[g/kg]	10-14	10-40	-
resp.	[g/Nm^3]	-	-	0.01 - 0.06
TOC load	[g/kg MSW]	2-3[1)]	0.1 - 1.0 [2)]	0.05 - 0.30 [3)]

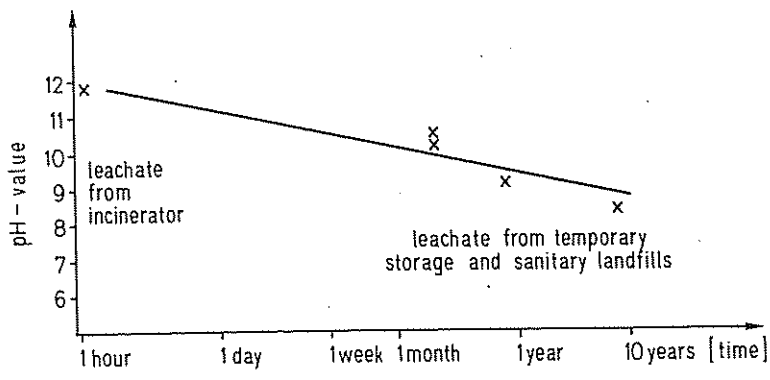


Fig. 1
pH-values of slag leachates as function of age of slag deposits: pH= 10.1- 0.28 ln t (p<1%); with t= age of slag in months.

Table 2
Concentration and load of TOC in filter dust samples from four Swiss MSW incinerators

MSW incinerator	Total Organic Carbon concentration [g/kg]	Total Organic Carbon load [g/kg MSW]
A	39	0.98
B	10	0.11
C	23	0.37
D	22	0.44

[1] Hutzinger, L., Olie, K. and Vermeulen, P.C., "Chlorodibenzo-p-dioxins and Chlorodibenzofurans are Trace Components of Fly Ash and Flue Gas of Some Municipal Incinerators in the Netherlands", Chemosphere, 6. 445-459 (1977)

[2] Brunner, P.H. and Moench, H., "The Flux of Metals through Municipal Solid Waste Incineration", Waste Management and Research, 4, 105-119 (1986)

[3] Brunner, P.H., Mueller, D.M., Moench, H., Mc Dow, S.R., "Total Organic Carbon Emissions from Municipal Incinerators", Waste Management and Research, 5, in press (1987)

[4] Hagenmaier, H., "Dioxine in Filterstäuben", 337. Dechema Kolloqium, April 2, 1987

GROUNDWATER FLOW IN GRAVEL DEPOSITS; INFLUENCE OF HIGHLY - PERMEABLE ZONES ON GROUNDWATER VELOCITY DISTRIBUTION

Peter Huggenberger, Christoph Siegentaler, Fritz Stauffer (ETHZ), Kerry Kelts

Fluvio-glacial gravel deposits make up most of the groundwater reservoir in Switzerland. At the outcrop scale these deposits display a complex pattern of sedimentary structures including channels, scour pools, chutes, overbank, drapes and clay seals. The divers structures comprise deposits with variable hydraulic conductivities. Horizontal- and crossbedding impart a direction dependency for these parameters. A minor part of these deposits is made up of stringer gravels without any fine fraction (layers "A" on fig. 1). The hydraulic conductivity of such zones is several orders of magnitude higher then in the surrounding sediments. The degree of interconnection of these layers and their extent determine the easiest flow path. Deterministic numerical models help visualize the influence of sedimentary structures on groundwater circulation at different scales.

Fig. 2 shows an idealized representation of the outcrop situation (fig. 1) into finite elements with their appropriate material properties (hydraulic conductivity, angle of anisotropy). For calculations, it was assumed that the

Fig. 1
Sedimentary-structures within fluvio-glacial gravel deposits of the Northern Swiss Midlands. (Pit Hüntwangen, Profile E-W)

A.)Well sorted gravel beds without fine-fraction (Openframe work zones)
B.) Sand lenses

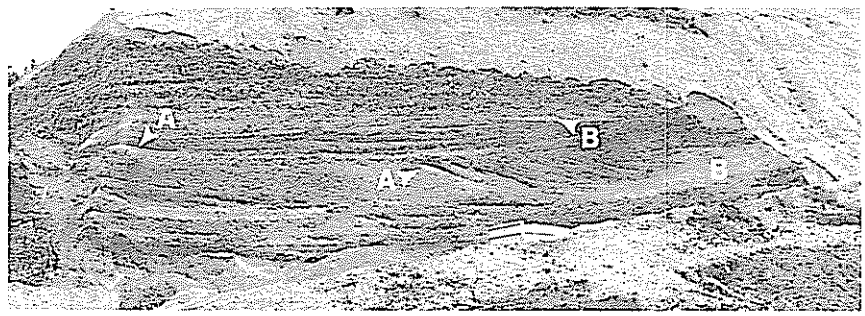


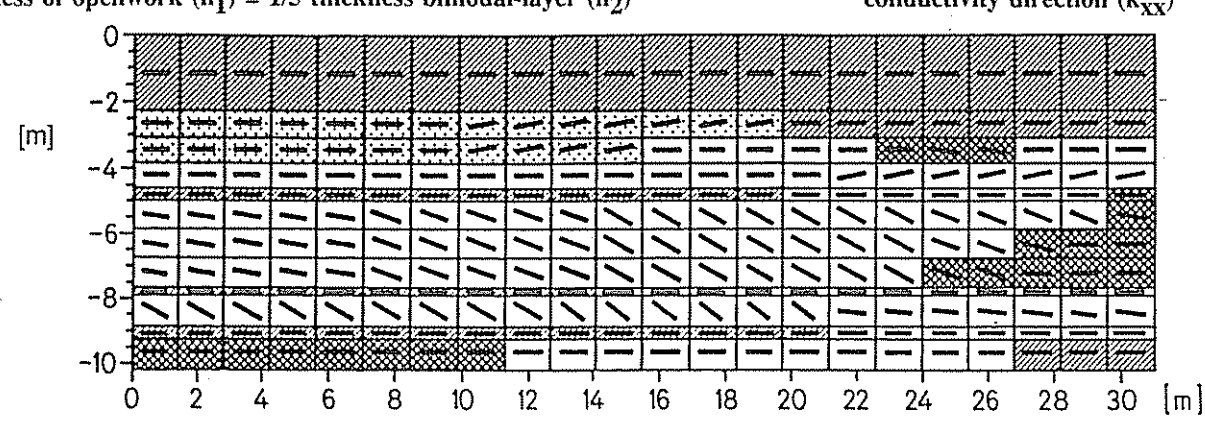
Fig . 2
Idealized representation of the outcropping sedimentary structures of the E-W profile (fig.1) as a finite element grid.

Composite sedimentary structure with openframework layers

Lithologies :	k_{xx}[m/s]	k_{yy}[m/s]
Boulder gravels (general trend of anisotropy about 0^0)	6×10^{-4}	1.0×10^{-4}
Grey-blue gravels with Openwork stringers (Material* taken as homogeneous but anisotropic; angle of anisotropy varies between 0^0 and 10^0)	7.0×10^{-4}	5.0×10^{-4}
Openwork/Bimodal couplets (homogeneous*, anisotropic; direction of anisotropoy variable)	8.0×10^{-2}	2.5×10^{-2}
Sand (angle of anisotropy variable)	7.5×10^{-5}	3×10^{-5}

* $k_{xx} = (\sum k_i h_i)/h_{tot}$; $k_{yy} = h_{tot}/(\sum k_i h_i)$;
thickness of openwork (h_1) = 1/5 thickness bimodal-layer (h_2)

maximum hydraulic conductivity direction (k_{xx})



potential gradient as well as the specific discharge through the block section profiles are constant. Top and bottom of the section are no-flow boundaries. Fig. 3 illustrates potential- and streamfunction contourlines for this simulation. A dense line pattern means higher groundwater velocities assuming constant porosity. The sand bodies (layers) slow the groundwater flow. Travel times of an input step-function (an abstraction of a tracer experiment; fig. 3) have been determined from streamfunction density pattern. Only advection (no dispersion and no molecular diffusion) have been considered in the calculation. The breakthrough curve (fig. 4) shows a rapid first appearence and a long tail due to the slower flow path. The differences in groundwater velocity, caused by the sediment structures and textures are of importance for questions dealing with the transport and mixing of pollutants in the groundwater.

This multidisciplinary project, together with the Institute of Hydromechanics at the ETHZ, is aimed at determining actual structural- and facies relationships among glaciofluvial gravel bodies in order to contribute further to realistic transport models of chemicals in groundwater.

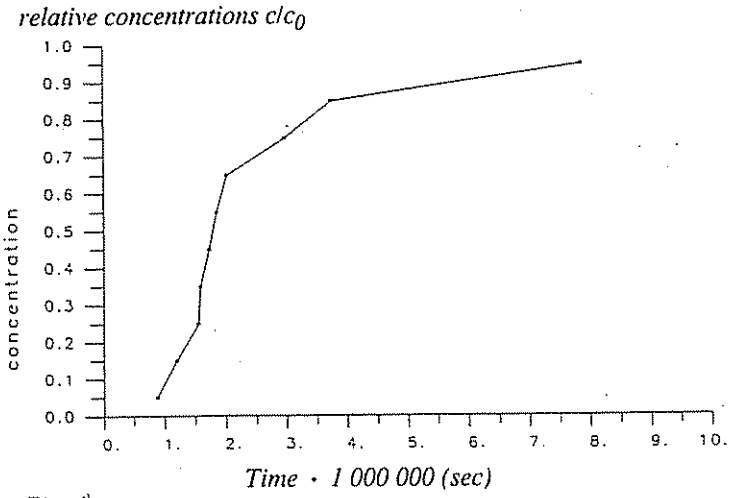


Fig. 4
Breakthrough curve of an input step function

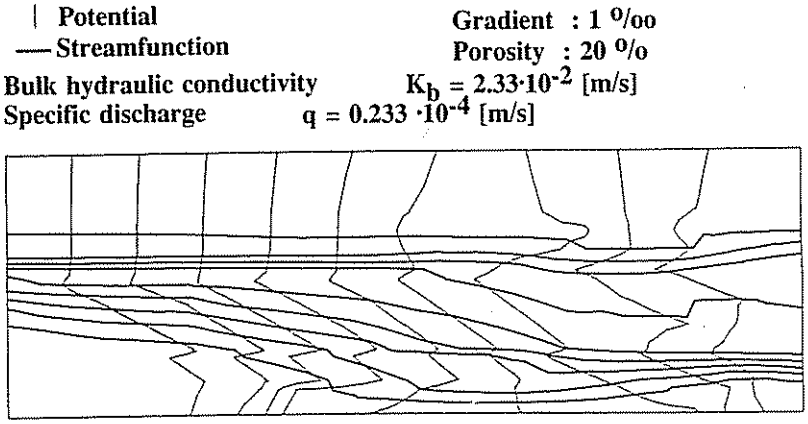


Fig. 3
Streamfunction and potential contour lines.
Boundary conditions:
A. Potential calculation: potential on left side: $\emptyset=\emptyset_0$, on right side: $\emptyset=\emptyset_1$.
B. Streamfunction calculation:
upper boundary $\Psi_1 = 0.23$ (L^2/T)
lower boundary $\Psi_0 = 0$ (L^2/T)
Dense streamline pattern means faster groundwater velocities assuming uniform porosities.

NEWS ABOUT EAWAG-COLLABORATORS

About the departure of Wolfgang Geiger

Dr Wolfgang Geiger, head of the Fisheries Section, retired end of July, 1986. For more than thirty years he was a pioneer in fishery research and established qualitative criteria for fish rearing in our waters. Wolfgang Geiger received his training in different places. He studied zoology at the University of Basle where he obtained his Ph.D. on the relative cerebral growth of bony fish under the direction of Prof. Portmann. Between 1952 and 1954 he was visiting research scientist at the Institut de pêches maritimes du Maroc in Casablanca. From 1954 to 1956 he worked at the Zoological Institute of the University of Berne, and in 1956 he joined the Swiss Federal Forestry, Wildlife and Fishery Board in Berne. Some of his most important work dates back to those days, particularly investigations on the stock and catch in trout rivers and on the reproduction biology of brown trout. This is when he gathered the necessary experience for his future activity as expert.

In 1962 he moved to the Zoological Institute of Geneva where he participated as senior scientist in the training of students and carried out, among other things, investigations on the sperm of salmonid fish. In autumn 1969 he followed Prof. Jaag's call to set up and head the newly established Fisheries Section at the EAWAG.

Wolfgang Geiger attached great importance particularly to the training of candidates for diploma and doctor's degrees. Thanks to his patience and insight of people's nature, he managed to impart, besides his fascination for fish, scientific thoroughness and objectivity. Thoroughness and realism characterize also his own work which covers, in a broad sense, the entire field of ichthyology: ichthyobiology, physiology, biochemical taxonomy, population biology, fish culture in our waters, effects of contaminants and technical interventions on the aquatic ecosystem.

Retirement is not an end, but a new beginning. All his friends, students and colleagues wish him all the best for this new beginning whose location is nowhere else than at the place of his youth, which thereby confirms the homing theory.

Dr Rudolf Müller

Perspectives of the Fisheries Section

The present time is characterized by an ever increasing number of rapid alterations of our environment. This situation leads the environmental scientist to reconsider the aim of his task from time to time in order to cope successfully with newly arising problems. Such a process of readjustment in the scope of activity includes the difficulty to identify the most urgent problems within the multitude of existing problems. The following issues are recognized today as being of prime importance:

- ecotoxicity of treated wastewater
- canalization of running waters/residual run-off in rivers
- lake eutrophication/oligotrophication

We have become aware of the general lack in understanding the role which fish play in the ecosystem of our waters. This has led us to define four main working areas. It is the common aim of these research efforts to contribute to the conservation of natural aquatic communities in an environment altered by man.

1. Ecotoxicology and fish

The mode of action of certain chemosynthetic compounds (or groups of compounds) on fish will be investigated physiologically, embryologically and ethologically. The first step in this direction at the Fisheries Section will be a doctoral thesis by T. Walter. The compounds investigated are representative of the innumerous organic compounds entering the rivers through treated wastewater effluents. We also hope to make use of fish as bioindicators for chemical interference at higher trophic levels of food-chains. The project is part of the EAWAG research focus "Ecotoxicology in Aquatic Systems" and will start in summer 1987.

2. Food-webs and nutrient turnover

The main question within this topic is: How do fish and food organisms interact? For lakes (Lake Hallwil, Lake Sarnen), the activities focus on the interdependence of zooplankton and fish at different trophic levels. For running water, on the other hand, the role of allochthonous versus autochthonous food for fish in River Glatt downstream of Lake Greifensee will be studied. This activity is situated within the EAWAG research focus "Running Waters". Started in March 1986, the feeding ecology of whitefish fry in lakes is currently being investigated in cooperation with D. Ponton, guest scientist from the Institut de Limnologie/INRA, Thonon (France).

3. Population ecology in running waters

During our investigations on River Rhone in Geneva in 1985/86 we realized how little is known about the fish populations and their natural fluctuations in large rivers. The fact that existing hydroelectric power plants and river bank constructions will have to be renewed in the near future, with additional and severe impacts on our rivers, makes it necessary to expand our basic understanding of the fish fauna in such waters. In the first place, the relationship between physical factors and population fluctuations due to migration, mortality and natural reproduction will have to be elucidated. Particular attention will be paid to the effects of river construction on fish.
The problem of residual (artificially reduced) water flow and its effects on stream ecology is becoming more and more acute. Hydropower development is intensifying its interest in small rivers which have remained untouched so far. Experimental evidence of such effects is badly needed in order to argue successfully for conservation of the river ecosystems.

4. Population ecology in lakes

One of the most advanced hydroacoustic systems available today enables us to quantify fish biomass and to determine length distributions of fish *in situ*. Activities are under way on Lake Hallwil (eutrophic) and Lake Sarnen (oligotrophic). We expect to make significant progress in understanding the reaction of the lake ecosystem - and in particular of fishes - to a changing stress such as the process of eutrophication/oligotrophication. The importance of this topic is reflected by a growing number of scientific events like workshops dealing with these questions.

Teaching, the second of the three groups of activity, is currently being intensified to meet the growing interest.

Consulting activities, on the other hand,will be restricted to projects contributing substantial new knowledge in fish biology and aquatic ecology. It should however be mentioned here that consulting has recently opened up access to certain research areas, set-ups and technologies otherwise inaccessible: Sedimentation in rivers due to reservoir evacuation, interesting but restricted fish waters, and advanced hydroacoustics. In fact, a continued exchange of ideas with people engaged in practical fishery is part of the scope of EAWAG and could prove to be essential for the identification of problems to come. Thus, the activities of the Fisheries Section during the next years can be summarized as: Priority on research based on clear concepts, complemented by selective consulting and transfer of new knowledge through teaching at university.

Dr Rudolf Müller

Promotion

Dr Willi Gujer has been promoted to Privatdocent for Water Technology and Water Protection at the ETHZ (Dept. of Civil Engineering). The title of his habilitation thesis is: A dynamic model for simulation of complex activated sludge systems. PD Dr Willi Gujer joined the EAWAG in 1974 and became head of the Engineering science department in 1977.

Member of parliament

The editor of the EAWAG news has been elected a member of the State (Canton) Zurich Parliament. As a member of the Green Party, Diana Hornung will have additional leverage for introduction of environmental ideas and concepts into legislation and practice. The Direction of EAWAG is very pleased about having one of its staff members in this position.

Death of Prof. Kurt Grob

Kurt Grob, professor at the Swiss Federal Institute of Technology Zurich and former research scientist at the EAWAG, died on 2 March 1987 at the age of 67.

Before joining EAWAG in 1974, Kurt Grob held a position as chemistry teacher at the grammar school and at the University of Zurich. Besides his teaching activities, he first worked in the tobacco industry where he studied the composition of tobacco smoke. This work brought him into contact with the analytical technique of gas chromatography which then became Kurt Grob's main field of research. During 25 years of intensive work he was able to greatly improve this methodology. Many techniques in the field of high-resolution gas chromatography are closely associated with Grob's name There are applications todayin many areas of analytical chemistry such as environmental chemistry, forensic science, food analysis and clinical chemistry.

After having established the considerable improvement achieved by the application of glass capillary gas chromatography for the determination of organic trace contaminants in water, he transferred his laboratory to the EAWAG. His analytical methods allow the quantitative determination of trace pollutants down to 10^{-9} g/l for individual organic chemicals in water. Kurt Grob also aided in promoting the application of capillary gas chromatography through his teaching at the University and at the Federal Institute of Technology in Zurich. He authored over 150 publications including a monograph on "Making and Manipulating Capillary Columns for Gas Chromatography". The University of Berne, as well as the ETH, awarded him a honorary doctoral degree for his outstanding achievements as a teacher and a scientist. Kurt Grob retired in summer 1985 after having completed, in his typically consistent manner, almost all of his research work.He died less than two yeasrs after his retirement. The EAWAG is deeply indebted to Kurt Grob and to his wife and assistant in the laboratory, *Mrs. Gertrud Grob*, for their unique scientific contributions.

Doctoral degree honoris causa

On June 4, 1987, the University of Geneva has awarded Prof. *Werner Stumm* a honorary doctoral degree for his outstanding achievements as chemist in the field of equilibrium chemistry of natural waters and their environment. By connecting theory and practice he developed mighty tools to help to better protect our environment.

8th Ozone World Congress and Exhibition

The 8th Ozone World Congress and Exhibition will take place in Zurich on 15-18 September 1987. It will be of particular interest to specialists in the field of technical ozone production, process engineering for waterworks, swimming pools, exhaust air decontamination, and water quality and to municipal authorities responsible for drinking water quality.

It is an honor for Switzerland to host this conference, because the Swiss waterworks, Swiss engineering companies and EAWAG have developed a tradition for ozone application and research.

A research group from EAWAG will report their results on the kinetics of ozone decomposition in water. Before the conference, EAWAG will host a workshop for chemists to discuss fundamental problems related to analysis of ozone and the formation of secondary products during water ozonation.

Environmental sciences as new type of study

For the first time, the ETHZ offers a 4 year study program in environmental sciences starting fall 1987. It is addressed to students with a broad interest in integrated natural sciences and requires strong motivation.

For questions refer to PD *Dieter Imboden*, EAWAG.

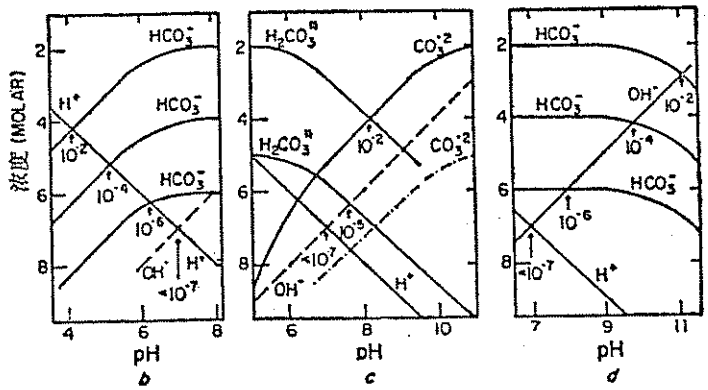


图 4.2 $CO_2(H_2CO_3^*)$、$NaHCO_3$ 和 Na_2CO_3 的纯溶液在不同稀释程度时的 pH (见例 4.1)。(a) 中的曲线可以用方程式 (12)—(16) 计算或借助于对数平衡图求得。(b—d) 图分别属于 CO_2、$NaHCO_3$ 和 Na_2CO_3 的纯溶液。对某些浓度，代表相应属于余件的那些交点用箭头指出。

点的pH值。例如纯 $NaHCO_3$ 溶液的质子余件是 $[H_2CO_3^*] + [H^+] = [CO_3^{2-}] + [OH^-]$。如图 4.2 c 所绘，在高浓度 ($C_T > 10^{-3}M$) 时，平衡可用简化的余件 $[H_2CO_3^*] \approx [CO_3^{2-}]$ 来表征。在此浓度范围内，$NaHCO_3$ 溶液保持一个恒定的 pH (由于浓度改变而产生的微小变化予以忽略)。在较稀的浓度范围里 ($10^{-4}M > C_T > 10^{-7}M$)，纯 $NaHCO_3$ 溶液的 pH 由其相应的质子余件 $[H_2CO_3^*] \approx [OH^-]$ 所表征。当 $C_T \to 0$ 时，电中性余件成为 $[H^+] \approx [OH^-]$，于是溶液呈中性 (pH ≃ 7)。CO_2 的纯溶液 ($C_T > 10^{-4}M$) 和 Na_2CO_3 的纯溶液 ($C_T > 10^{-3}M$) 的 $d\mathrm{pH}/d\log C_T$ 值分别为 − 0.5 和 + 0.5。

Chinese edition of Aquatic Chemistry, 1987

The second edition (1981) of the book "Aquatic Chemistry - An Introduction Emphasizing Chemical Equilibrium in Natural Waters" has been translated under the supervision of Prof. Tang Hongxiao of the Institute for Environmental Chemistry of the Chinese Academia Sinica. He spent many years in the USA and one year at the EAWAG. Thanks to his skill, non-translatable expressions, such as the names of the authors, Werner Stumm, and James J. Morgan, retain the English pronunciation, although still expressed in Chinese characters.

Investigations on the toxicological effects caused by the fire at Schweizerhalle on the Rhine

After the great fish kill and the initially only suspected important damage caused to the Rhine's ecosystem downstream of Schweizerhalle, the EAWAG was commissioned by the governement of the canton Basle-Land to make an inventory of the damage and to develop measures for the regeneration of the Rhine.

The EAWAG divided this mandate into three parts:

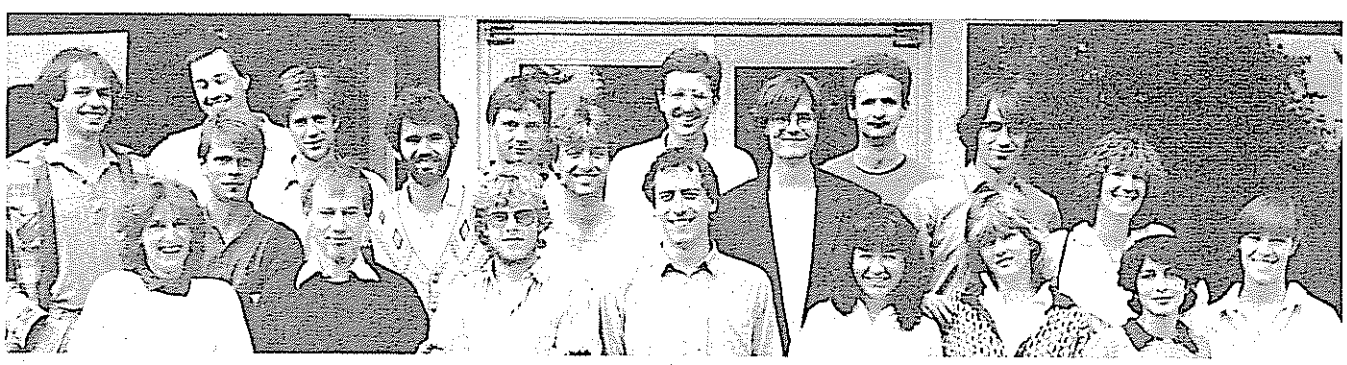
1. Chemical and biological inventory in the flowing waters, in the benthos and in the sediments.
2. Determination of the chemodynamic behavior of the chemical substances discharged into the Rhine with the fire extinguishing water and assessment of their ecotoxicological effects.
3. Development of measures for the regeneration of the Rhine, primarily with fish, and monitoring of the restocking measures of the Rhine with organisms.

A first intermediate report on the primary effects of the Rhine's contamination with mercury and thiophosphoric ester compounds was delivered already 4 weeks after receipt of the mandate. The investigations revealed that the macroinvertebrates had been almost entirely destroyed while the fish populations of eel and grayling had been totally wiped out. Only the aquatic plants did not suffer any important damage. The microbiological population structure was intact during our investigations about three weeks after the accident. The Rhine's self-purifying capacity was not impaired at the time of these investigations. Nevertheless, a significant biodegradation could not be expected for many discharged substances during the flow time.

Our current investigations on the chemical substances are trying to establish, with the help of chemodynamic measures, why the compound concentrations in the Rhine had been so terribly toxic. From theses analyses we expect to obtain important information on the behavior of the abiotic substances in aquatic systems. The field studies primarily aim at restocking the Rhine with aquatic organisms. Since no chemical residues were found on the river bed along the Rhine section at Basle-Stadt, some species of invertebrates are expected to reappear even this year. However, on account of the still prevailing too high load of abiotic compounds of different origins in the Rhine, the creation of a natural biocenotic habitat is not foreseeable in the near future. The availability of sufficient fish food organisms will soon enable important fish restocking measures.

Dr Peter Perret

Participants of the seventh postgraduate course in Sanitary Engineering and Water Pollution Control (offered by the Institute for Water Pollution Control, IGW, and the Institute for Hydraulics and Water Resources Management, IHW, of the ETHZ).



From left to right, front row: *Renate Krauss, Jakob Hedegaard, Patrick Höhener, Haroun Frick, Judith Kemmler, Slavica Prgomet, Mahnaz Schneeberger, Lea Locher-Azevedo;*
second row: *Georg Furger, Urs Dietschi, Jürg Kappeler, Marcel Fisch, Thomas Walter, Nancy Dise;*
third row: *Paul Wersin, Manuel Elgorriaga, Martin Anderson, Thomas Wepf, Urs Kempf, Herbert Kessler.*

In 1986 and 1987, the EAWAG was honored by the visit of the following **guest scientists**:

Allen Evelyn, Biologist, Univ. of Michigan, An Narbour, USA, (June-Aug. 86).
Bruno Jorge, Chemist, Royal Inst. of Techn., Stockholm, Sweden (May-July 87)
Capri Silvio, Chemical Engineer, Ist. di Ricerca sulle Acque, Rome, Italy, (Jan.-June 86 and June 87).
Christensen Erik, Ass. Prof. Civ. Eng., Univ. of Wisconsin, Milwaukee, USA, (June-Aug. 86).
Czuczwa Jean, Dr., Analytical Chemist, Indiana Univ., Bloomington, USA, (Jan.-May 86).
Drever James, Prof. of the Univ. of Wyoming, USA (Sept 87-Sept.88)
Emerson Steve, Prof. Univ. of Washington, Seattle (Feb.-Sept. 87)
Fuscoe Wendy Lynn, North Carolina State Univ., Chapel Hill, USA, (Sept.-Dec. 86).
Goncalves Maria de Lurdes, Prof., Centro de Quimica Estrutural, Inst. Superior Technico, Lisboa, Portugal (July-Aug. 87)
Heyerdahl Emily, Geologist, Oregon Graduate Center, Beaverton, USA, (May-Oct. 86).
Johnson Carola, PhD, Chemist, Imperial College, Geology Dept., London, UK, (until March 86).
Lerman Abraham, Prof., Chemist, Northwest Univ. Chicago, USA, (Nov. 86).
Marcomini Antonio, Dr., Chemist, Dip. di Scienze Ambientali, Univ. Venezia, I, (May- June 86 and 87).
Masten Susan, Engineer, Harward Univ., Cambridge Mass., USA, (until April 86).
O'Connor John, Prof., Univ. of Missouri- Columbia (July-Aug. 87)

Pavlova Violeta, Chemist, Univ. Göteborg, Sweden, (Sept. 86- Sept 87).
Pollinger Utsal, Prof., Israel Oceanographicand Limnol. Res. Inst., Haifa, Israel (Aug.-87-July 88)
Ponton Dominique, Biologist, Inst. de Limn., Thonon, France (Feb-Dec 87)
Pytkowicz Richard, Prof. Oregon State Univ., Oregon, USA, (Jan.-March 86).
Rebhun Menahem, Prof., Technion, Haifa, Israel (July-Dec 87)
Stephanou Euripides, Prof. Univ. Heraklion, Greece (June-Aug. 87)
Sridhar Mynepalli, Prof., Engineer,Dept. Preventive and Social Medicine, Univ. of Ibadan, Nigeria, (Oct. 86- Sept. 87).
Wang Ao Sheng, Geologist, Inst. of Geography, Academia Sinica, Nanjing, China (Feb. 87-Feb 89)
Xue Hanbin, Chemist, Inst. of Environ. Chemistry, Academia Sinica, Beijing, PR China, (March 86- March 87).
Yu Jun Qing, Geologist, Qing Hai Salt Lake Res. Inst., Academia Sinica, Xinhing, PR China, (Sept. 85-Sept 87).
Zepp Richard, Prof., Univ. of Miami, USA (June- Aug. 87)

The **Otto Jaag Prize 1986** for the most outstanding thesis in the field of water resources and water pollution control has been awarded to *Gerhard Furrer* for his thesis on the surface controlled dissolution of metal oxides.

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Nearly hundred participants discussed at a workshop the possibilities of lake restoration. The speakers and discussion chairpersons were from left to right: Prof. Bernhardt, Dr Imboden, Prof. Vollenweider, Dr Davis, Dr Gächter, Prof. Lerman, Prof. Forsberg, Dr Davison, Prof. Bachofen, Dr Perret, Dr Sigg, Dr Clasen, Prof. Stumm, Dr Reynolds, Dr Pechlaner. The proceedings of this workshop have been published in a special issue of the Swiss Journal of Hydrology.

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