特別講演

特別講演 1	H. R. von Gunten
特別講演 2	J. Alstad

Cyclic metal migration in a groundwater stream

by H. R. von Gunten
Laboratorium für Radiochemie, Universität Bern, Switzerland
and
Paul Scherrer Institut, Würenlingen, Switzerland

Abstract:

Alpine and peri-alpine countries recover drinking water to a large extend from shallow groundwater resources in widely distributed glacio-fluvial deposits. The groundwater in these quaternary sediments is mainly recharged by infiltrating river water. Due to the importance of this process for the water quality it is mandatory to understand the biological, chemical and physical changes which occur during infiltration and movement of water in aquifers.

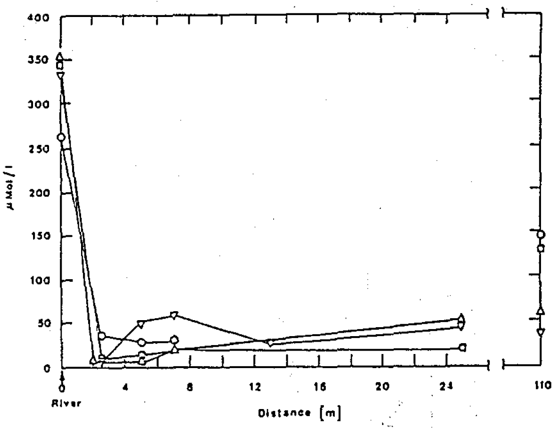
In the present study we concentrate on daily, annual and spacial changes of some dissolved inorganic water components and other properties in the River Glatt (Switzerland) and in the adjacent groundwater stream.

A well equipped study site is available for these investigations. Here, water from the polluted River Glatt infiltrates continuously with a mean linear flow velocity of 2-4 m d^{-1} through the saturated and stratified ground zone and becomes the uppermost layer of the local groundwater stream.

Diurnal variations of several parameters (e.g. pH, O_2, Al) were observed in the river water. These systematic fluctuations are much larger in summer than in winter and are accentuated during sunny days. They are the result of photosynthesis and respiration of aquatic biota. Upon infiltration in the ground, these variations are almost completely damped due to chemical reactions involving $CaCO_3$ dissolution/precipitation. However, very drastic decreases in pH and dissolved oxigen (Fig. 1) are observed during the infiltration of river water into the aquifer. These sudden changes in the infiltrating water influence solu-

bilities and redox properties of several chemical elements. The chemical changes during infiltration of river water can be described fairly well by a few simple electron transfer (involving organic matter degradation, denitrification, nitrification) and weathering reactions. Of prime importance during the warm summer months is the oxidation of organic matter and the denitrification process. Both reactions are strongly influenced by temperature enhanced micro-biological activities.

Figure 1

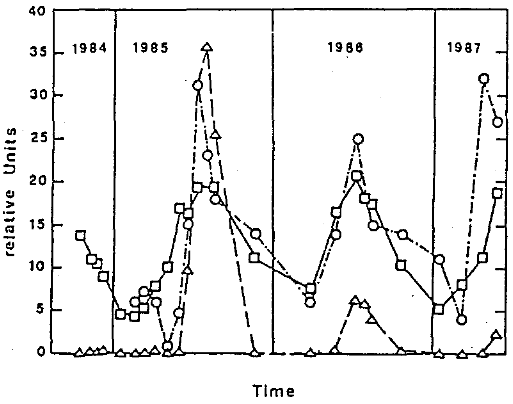


Changes in the concentration of dissolved oxygen during infiltration of river water in 1985 and 1986. The concentrations in the river are shown at distance zero. Notice a general increase in oxygen concentrations at greater flow distances in the aquifer. They are produced by admixture of deeper, oxygen-rich groundwater. □ July 1985, △ August 1985, o August 1986, ▽November 1986.

The annual temperature variations in river and groundwater are responsible for biology- and solubility-related cyclic changes in many of the water parameters. Most of these changes in the groundwater are highly correlated (e.g. pH, Si-, Mn-, Cd-concentrations) or anticorrelated (e.g. alkalinity, O_2, NO_3^-, Ca^{2+}) with the water temperature.

The enhanced degradation of organic matter at higher temperatures during summer leads to a larger consumption of oxygen and, hence, to a reduction in the redox potential which becomes low enough to cause the dissolution of manganese oxides and hydroxides. The redox potential was, however, never low enough to cause iron oxides/hydroxides to dissolve. The annual changes in redox potential lead to very pronounced Mn^{2+}-peaks in the groundwater during summer time (Fig. 2). No such peaks are observed in the river water which is, therefore not the source for the high Mn^{2+}-concentrations in the groundwater stream. Recent measurements in the pore-water of Glatt River sediments have shown Mn^{2+}-concentrations . 500 ppb in summer 1987. Thus, the sediments in the river bed can be considered as a source for the dissolved Mn. Additional Mn^{2+} may result from the dissolution of aquifer material during reducing conditions. The magnitude of the Mn-peaks varies from year to year and depends on the climatic conditions. The dissolved Mn^{2+} does not migrate very far in the groundwater field; it is soon reprecipitated as manganite.

Figure 2



Annual variability of temperature °C,□; Mn^{2+},△ (one unit = 10 ppb); Cd^{2+},○ (one unit = 0.01 ppb) at the well in 5 m distance from the River Glatt. Notice different scales for Mn^{2+} and Cd^{2+}.

Fig. 2 also demonstrates that the concentrations of cadmium in the groundwater are highly correlated with those of Mn^{2+}. Again, the Cd-peaks show no relation to river concentrations. We assume that the chemical behaviour of Cd is influenced by co-dissolution/-precipitation of Mn or $CaCo_3$. A possible additional source for Cd may be degraded organic material. Cadmium also disappears, like Mn, after relatively short migration distances.

Fig. 2 shows a very high correlation of Mn and Cd with water temperature and, thus, with bioactivity. Material balances suggest that the observed concentrations (not shown here) for Cu and Pb result from the degradation of aquatic biota, whereas Zn-concentrations are mostly influcenced by river water concentrations.

The infiltration of dissolved, particulate and colloidal compounds from the river Glatt into the groundwater was further investigated, using the fallout of the reactor accident at Chernobyl. The radionuclides ^{103}Ru, ^{131}I, ^{132}Te, $^{134,137}Cs$ were measured several times in the river and in the groundwater. Dissolved (> 0.05 µm) ^{131}I and ^{103}Ru infiltrated from the river into the groundwater practically without delay. ^{132}Te was retarded by the aquifer material and ^{134,137} Cs was not observed in the groundwater. This behaviour of Cs was expected, based on batch and column sorption experiments performed in the laboratory using fractions of the aquifer material. Particulate and colloidal infiltration of radioactivity from the river into the groundwater is a slow process of minor importance.

NEW DEVELOPMENTS IN THE SISAK TECHNIQUES FOR STUDYING SHORTLIVED NUCLIDES.

J. Alstad, The Göteborg-Mainz-Oslo SISAK-collaboration.

Abstract

A review of the recent experimental investigations based on the SISAK techniques of studying nuclear decay properties is given. The main efforts have been devoted to developing fast chemical separations of Tc, Ru, Rh and Pd and measuring γ-energy values and transitions in β-decay of neutron-rich isotopes of these elements.

The most neutron-rich nuclides observed so far are ^{109,110}Tc, ^{109,111}Ru and ^{113,115}Pd which have been investigated by measuring single γ-ray and $\gamma\gamma$ (t)-coincidence spectra.

The chemical separation procedures are sketched with emphasis on the very fast chemistry.
In the investigated region the level schemes may be explained to consist of two different types of energy states, proton-holes connected to a sperical shape nucleus and proton particles with rotational band-type structure of a deformed nucleus. The energy value in a level of the latter for odd mass number isotopes varies regularely with mass number and passes a minimum midway between the neutron number N=50 and N=82.
$\gamma\gamma$-coincidence measurements of shortlived nuclides require high flow-rates of liquids in the solvent extraction separation steps over sizable periodes of time. A need for a reduction in the volumes of the liquide wastes has been a primary reason for a development of new centrifuges with internal volumes less than 1 ml. The new separation system consists of
1) a degassing unit, which traps aerosol clusters in small liquid volumes from the gas-jet and separates the carrier gas and the liquid in a gas-liquid centrifuge and
2) centrifuges for separating liquid mixtures in the 1 ml volume range. The centrifuges are running at a speed of up to 45 000 r.p.m.
The construction and properties of the new centrifuges and their applications is discussed.