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# Large-scale redox plume in glaciofluvial deposits due to sugar-factory wastes and wastewater at Aarberg, Switzerland

P. Wersin · J. Abrecht · P. Höhener

**Abstract** The sugar factory at Aarberg, Switzerland, has processed about  $18 \times 10^6$  metric tons of sugar beets in the last 100 years. This has been accompanied by releases of dissolved organic carbon to the groundwater, induced both by direct wastewater disposal until 1964 and by ongoing leakage from solid-waste deposits. Downgradient in the groundwater of the glaciofluvial aquifer, depletion of oxygen concentrations accompanied by low nitrate, high ammonium, dissolved Mn(II) and Fe(II) concentrations are observed. This study was aimed at developing a quantitative comprehension of the impact of the leaking waste deposits on biogeochemical processes in the aquifer and on groundwater quality. The study includes a review of historical information, a survey of the hydrogeochemistry in the aquifer, the characterisation of river-water infiltration rates with the radon method, establishment of a mass-balance model based on a numerical flow and transport model, and application of a stable-carbon-isotope method to show biodegradation of sugar-waste deposits in the aquifer. The investigations demonstrate that present emissions from waste deposits would not lead to the consumption of all the  $O_2$  in the aquifer. The present occurrence of anoxic groundwater conditions is explained as a result of the long history of waste loading.

**Résumé** La sucrerie d'Aarberg (Suisse) a traité environ  $18 \times 10^6$  tonnes de betteraves à sucre au cours des 100 dernières années. À cette activité sont associés des rejets

de carbone organique dissous dans la nappe, produit à la fois par le stockage direct d'eaux usées jusqu'en 1964 et par les fuites continues à partir des dépôts de déchets solides. En aval, dans l'eau de l'aquifère fluvio-glaciaire, on observe une diminution des concentrations en oxygène associée à des concentrations faibles en nitrates, fortes en ammonium, en Mn(II) et en Fe(II) dissous. Cette étude a été réalisée pour disposer d'une connaissance quantitative de l'impact des fuites provenant des dépôts de déchets sur les processus biogéochimiques dans l'aquifère et sur la qualité de l'eau souterraine. Elle comprend une revue des informations historiques, un relevé hydrogéochimique de l'aquifère, la caractérisation des taux d'infiltration entre la rivière et la nappe par la méthode du radon, l'établissement d'un modèle de bilan de matière basé sur un modèle d'écoulement et de transport, et l'application de la méthode des isotopes stables du carbone pour montrer la biodégradation des déchets de la sucrerie dans l'aquifère. Ces recherches démontrent que les émissions actuelles à partir des déchets stockés ne permettent pas d'expliquer la consommation de tout l'oxygène au sein de l'aquifère. La présence actuelle de conditions anoxiques dans la nappe est expliquée par la longue histoire des apports de déchets.

**Resumen** La factoría de azúcar de Aarberg (Suiza) ha procesado alrededor de  $18 \times 10^6$  toneladas de remolacha de azúcar en los últimos 100 años. El proceso ha implicado la emisión de carbono orgánico disuelto a las aguas subterráneas, tanto por vertido directo de aguas residuales (hasta 1964) como por el lixiviado continuo de los depósitos de residuos sólidos. Aguas abajo, en las aguas subterráneas del acuífero glaciofluvial, se observa la ausencia de oxígeno, junto con concentraciones bajas de nitrato y altas de amonio, Mn(II) y Fe(II) disueltos. El objetivo del presente estudio consistía en cuantificar el impacto de los lixiviados de residuos sólidos en los procesos geoquímicos del acuífero y en la calidad de las aguas subterráneas. El estudio incluye una revisión de la información histórica, una campaña hidrogeoquímica, la caracterización de las tasas de infiltración del agua de río mediante el radón, el establecimiento de un modelo de balance de masas que está basado en un modelo numérico de flujo y transporte, y el uso de los isótopos estables del carbono para evidenciar la biodegradación de los depósitos de residuos de azúcar en el acuífero. Las investi-

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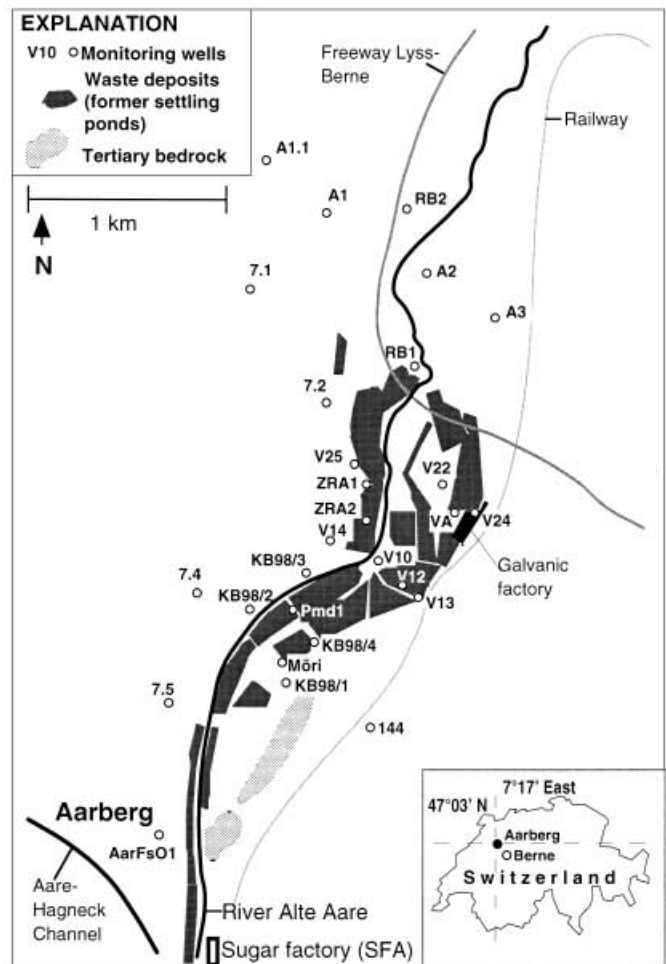
gaciones demuestran que las emisiones actuales de los depósitos de residuos no acarrearían el consumo de todo el oxígeno del acuífero. Por tanto, las condiciones anóxicas actuales de las aguas subterráneas se explican como consecuencia de un largo historial de aportación de residuos.

**Keywords** groundwater · pollution · redox plume · infiltration · stable-carbon isotopes

## Introduction

The preservation of groundwater quality in shallow porous aquifers such as glaciofluvial deposits is of great importance for the sustainable development of various regions. In recent years, an increasing number of studies reported on large-scale deterioration of redox conditions in shallow aquifers where groundwater would be naturally oxidic: such large-scale redox plumes were studied in detail and reported to originate from landfills (Reinhard et al. 1984; Lesage et al. 1990; Lyngkilde and Christensen 1992; Bottrell et al. 1995; Amirbahman et al. 1998) or from wastewater infiltration operations (LeBlanc et al. 1991). A few additional studies reported the impact of sugar mills and sugar refineries on regional groundwater quality (Schenk 1979; Ghio et al. 1985; Pawar et al. 1998). Their negative impacts on the groundwater quality were reported to result from the infiltration of sugar-refining-derived wastewater into groundwater, and from leachates formed from the disposal of solid wastes. The washing process of sugar beets, prior to refining, results in a suspension, highly enriched in organic carbon, which is often deposited in settling ponds. Compounds that can be leached from such settling ponds and that affect groundwater quality are dissolved organic carbon (DOC), ammonium and potassium, which originate from the sugar beets themselves or are by-products in the refining process (acids, bases, salts).

The sugar factory at Aarberg (SFA), founded in 1899, has been processing as much as 500,000 metric tons of sugar beets per year and is an important factor in the regional economy. The factory is located (see Fig. 1) in the Bernese Seeland, overlying one of the large gravel aquifers in the Alpine Foreland Basin (Biaggi et al. 1994). The aquifer consists of unconsolidated outwash deposits of glaciofluvial gravel and sand and is an important source for the regional drinking water supply. In the 1930s, it was first reported that groundwater quality downgradient of the zones where sugar beets were processed was deteriorating (Wersin and Abrecht 1999). Low oxygen concentrations in the groundwater, accompanied by high concentrations of ammonium and dissolved Mn(II) and Fe(II) were observed in several wells downgradient of areas of river infiltration and from leaky settling ponds. Optimisation of the refining process from 1964–1989, as well as the installation of wastewater treatment facilities and the end of solid residue deposition, were intended to combat the negative



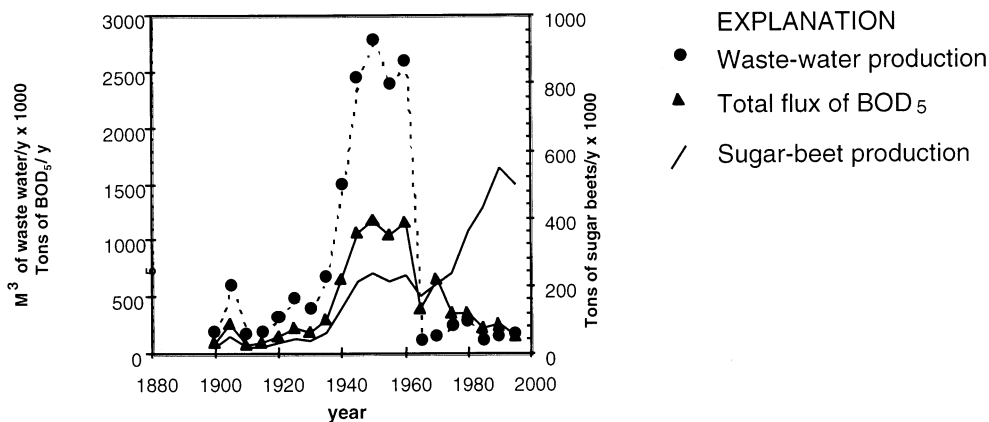
**Fig. 1** Map of the region underlain by the Bernese Seeland aquifer, showing locations of monitoring wells and sugar waste deposits (former settling ponds)

impacts on groundwater quality. However, a series of old leaky waste sites, where solid and liquid residues of the beet-refining process had been disposed of, still threaten the groundwater quality. Since 1960, a number of investigations have shown that the groundwater quality has improved in some monitoring wells, whereas it remained unchanged in others (summary in Wersin and Abrecht 1999). A comprehensive study of the impact of former waste deposits and infiltration ponds on groundwater quality is still lacking.

The motivation of this study was to provide a basis for decision-making with regard to monitoring and/or remediation measures. In order to predict the future development at a regional scale, a quantitative understanding of the biogeochemical processes in the aquifer is required. The study focused on answering the following questions:

1. What are the hydrogeological boundary conditions of the area affected by sugar-waste disposal?
2. Which are the most important biogeochemical processes in this aquifer, and how do the waste sites interfere with these processes?

**Fig. 2** Historical data on wastewater production, total flux of BOD<sub>5</sub> in wastewater and processing of sugar beets at the sugar factory at Aarberg



3. What are the present mass fluxes of the waste material in comparison with past fluxes?
4. Which fluxes of oxidants would be needed to keep the groundwater aerobic?

In order to answer these questions, the following work was performed:

1. Review of historical information with regard to past effluents from the sugar factory and past hydrogeochemical conditions in the aquifer.
2. Survey of the present hydrogeochemistry of the aquifer and the present effluents from sugar waste sites.
3. Characterisation of river-water infiltration rates with the radon method.
4. Application of a method, based on stable-carbon-isotope data, to show biodegradation of sugar-waste deposits in the aquifer.
5. Establishment of a mass-balance model based on a numerical flow and transport model.

The results presented in this study serve as a comprehensive database for further planning on a regional scale.

## Field-Site Description

### History of Waste Disposal

The manufacturing of sugar from sugar beets gives rise to high amounts of organic-rich wastewaters and sludges. The water consumption is about 10–15 m<sup>3</sup>/ton of manufactured beets, if water use is not optimised (Hoffmann 1985). The SFA processed around 50,000 tons of beets per year for the first 40 years (Fig. 2). Later, the sugar production was significantly increased and has reached over 500,000 tons per year since the 1990s. Parallel to this evolution, the amount and organic load of the wastewater increased. These relationships are shown in Fig. 2, where the degradable organic load is given as biological oxygen demand within 5 d (BOD<sub>5</sub>).

During the history of the SFA, two waste periods can be distinguished:

1. From 1899 to 1964 all of the sediment-rich wastewater was discharged to settling ponds located close to

the River Alte Aare (Fig. 1). These ponds were not equipped with impermeable liners and have to be considered as leaky. The remaining water was infiltrated in adjacent areas and to some extent conducted directly into the river. By deposition of the organic-rich sediment, the area was transformed into fertile land which could be used for agriculture. On the other hand, the discharge of large amounts of wastewater caused serious degradation of the underlying groundwater, involving the change from aerobic to anaerobic conditions. The highest amounts of discharged wastewater (around 1,000 t of BOD<sub>5</sub> per year) occurred within the period 1940–1964 (Fig. 2).

2. Since 1965 the quantity of wastewater and its release to the hydrosphere has significantly decreased (Fig. 2) by continuously improved wastewater management. The main improvements have included recycling of wash- and rinsing water (since 1965), the biological treatment of the wastewaters (since 1968) and the construction of leak-resistant wastewater reservoirs (since 1989) for the storage of pre- and post-treated waters during the intense sugar-processing season between October and November each year.

### Hydrogeological Setting

The waste deposits (former settling ponds) are located on the Bernese Seeland aquifer, which is bordered by two subparallel ranges of hills to the east and the west. The study area, which is intersected by the River Alte Aare (Fig. 1), is covered by forest and agricultural land. The aquifer is composed of Quaternary gravel-rich sand and irregular interlayers of fine-grained silty deposits (Kellerhals and Tröhler 1976). Below these glaciofluvial deposits fine-grained lodgement till and marls of the Tertiary Molasse form the aquitard. The aquifer dips from the easterly border of the valley to the west, resulting in an increase of aquifer thickness from 0 to 40 m in the centre of the valley.

The aquifer is unconfined and characterised by a shallow depth of the water table of 3–5 m, and high, variable hydraulic conductivities in the range of 10<sup>-4</sup> to 10<sup>-2</sup> m/s in the study area (Biaggi et al. 1994). Within the study

area the groundwater is recharged by river infiltration, groundwater flow from the eastern hill area, inflow of groundwater from the south and by precipitation. The flow regime varies seasonally, showing the highest flow occurring in the summer. In the study area, 28 monitoring wells were installed (Fig. 1). They were generally equipped with screens over the whole saturated extent of the aquifer above the aquitard, between 1.5 and 18 m.

### Field and Laboratory Methods

#### Groundwater sampling techniques

Groundwater samples were collected using downhole submersible electrical pumps (either Grundfos MP1, Bjerringbro, Denmark, or Whale Superline 991, Munster Simms Engineering, Bangor, UK). At least two well volumes were removed before samples were taken. Oxygen concentration, temperature and pH were determined immediately at the site using the appropriate electrodes (O<sub>2</sub> and temperature: Clark type electrode, WTW Oxi 96; pH: WTW 95; all from WTW, Weilheim, Germany) fitted to a flow cell. Samples for analysis of dissolved species were filtered in the field immediately after collection using 0.45- $\mu$ m polyvinylidene-fluoride filters (Millipore, Bedford, USA). Samples for analysis of cations and dissolved metals were acidified with 0.1% distilled concentrated HNO<sub>3</sub>; samples for S(-II) analysis were fixed using zinc acetate solution (Clesceri et al. 1989). For CH<sub>4</sub> analysis, 58-ml serum bottles were filled and closed without head space using butyl rubber stoppers. Within 24 h after sampling, a 10-ml head space of N<sub>2</sub> was introduced simultaneously with the removal of 10 ml of liquid. Gases were measured in the resulting head space after shaking followed by 4 h of equilibration at 7 °C. For stable-carbon-isotope analysis of the dissolved inorganic carbon (DIC), glass bottles (300 ml) were filled with unfiltered groundwater and closed without head space using rubber stoppers.

#### Infiltration measurements using the <sup>222</sup>radon method

The infiltration rates of water from the River Alte Aare into the aquifer were estimated using the method described by Hoehn and von Gunten (1989). Briefly, groundwater was collected from the upper, high-perme-

ability zones in the monitoring wells that were pumped by two pumps simultaneously, to obtain a flow divide between the upper and lower pump. The radon activity was measured on site using an electrostatic method (Rad7, Niton Inc., USA) as described in Hunkeler et al. (1997). From the radon activities, the radioactive decay constant of <sup>222</sup>Rn (0.18 d<sup>-1</sup>) and the equation described in Hoehn and von Gunten (1989), mean residence times ( $\tau$ ) of the groundwater between the river and the monitoring wells were calculated. Specific infiltration rates ( $q$ ) were obtained as described in Table 1.

#### Laboratory leachate experiments

Leaching experiments were performed in the laboratory for representative solid samples from different waste deposits of various ages. The goal was to estimate the composition of the leachates and the potential impact of the waste material on groundwater. Representative samples (100 g) from various waste deposits, obtained by drilling, were diluted in distilled water in a ratio of 1:10, and reacted for 24 h under air atmosphere. The water phase was subsequently filtered and analysed for DOC, K<sup>+</sup>, Na<sup>+</sup>, SO<sub>4</sub><sup>2-</sup> and chemical or biological oxygen demand (COD, BOD<sub>5</sub>). The solid material was analysed for water content, total organic carbon (TOC), COD and carbonate content.

#### Chemical analysis

Concentrations of Cl<sup>-</sup>, NO<sub>2</sub><sup>-</sup>, NO<sub>3</sub><sup>-</sup>, SO<sub>4</sub><sup>2-</sup>, PO<sub>4</sub><sup>3-</sup> as well as NH<sub>4</sub><sup>+</sup>, Na<sup>+</sup>, K<sup>+</sup>, Ca<sup>2+</sup> and Mg<sup>2+</sup> were determined by capillary electrophoresis (BMG Engineering Schlieren, Switzerland). Dissolved Fe and Mn were quantified by atomic absorption spectroscopy. It was assumed that the dissolved Fe and Mn consisted mainly of dissolved species with the redox state +II. S(-II) was measured colorimetrically according to standard methods (Clesceri et al. 1989) and represents the sum of H<sub>2</sub>S, HS<sup>-</sup> and S<sup>2-</sup>. Alkalinity was measured by potentiometric titration. The head-space technique was used to quantify CH<sub>4</sub> as described elsewhere (Bolliger et al. 1999). DIC concentrations were calculated from alkalinity and pH data (Stumm and Morgan 1996). DOC, COD and BOD<sub>5</sub> were measured using standard methods (Clesceri et al. 1989).

**Table 1** Determination of infiltration rates using the <sup>222</sup>radon method. *n.d.* Not determined, because distance from river was too large

Well	<sup>222</sup> Rn activity (Bq/L)	Residence time ( $\tau$ ) (d)	Distance to river $x$ (m)	Specific infiltration rate ( $q$ ) <sup>a</sup> [m <sup>3</sup> (m <sup>-2</sup> d <sup>-1</sup> )]
KB98/4	13.0	>15	310	Reference for no infiltrate
V10	7.1	4.4	20	0.68
KB98/2	5.5	3.1	25	1.21
KB98/3	8.0	5.3	40	1.13
RB1	5.6	3.1	50	2.47
ZRA1	7.6	4.9	80	2.67
ZRA2	7.2	4.9	80	2.40
7.5	12.0	14.5	330	n.d.
7.4	10	8.2	310	n.d.
RB2	8.8	6.3	170	n.d.
Mean				1.76 $\pm$ 0.85

<sup>a</sup> Specific infiltration rate ( $q$ ) was obtained using equation  $q = \phi \times x / \tau$ , where  $\phi$  is porosity (0.15),  $x$  is distance between river and monitoring well, and  $\tau$  is mean residence time

### Stable-carbon-isotope analysis

All measured  $^{13}\text{C}/^{12}\text{C}$  ratios are reported in the delta notation ( $\delta^{13}\text{C}$ ) referenced to the Vienna PeeDee Belemnite standard (Coplen 1996). The  $\delta^{13}\text{C}$  value is defined as:

$$\delta^{13}\text{C} = \frac{R_{\text{sample}} - R_{\text{standard}}}{R_{\text{standard}}} \times 1000 \quad (1)$$

where  $R_{\text{sample}}$  and  $R_{\text{standard}}$  are the carbon-isotope ratios in the sample and the standard, respectively. For stable-carbon-isotope analysis, the DIC was precipitated as  $\text{BaCO}_3$  within 24 h after sampling by addition of 3 ml of a  $\text{CO}_2$ -free 2-M NaOH and 10 ml of a  $\text{CO}_2$ -free 1.2-M  $\text{BaCl}_2$  solution. After more than 12 h of equilibration, the precipitate was filtered under a stream of  $\text{N}_2$  and dried at 105 °C for 12 h, the dried  $\text{BaCO}_3$  was converted to  $\text{CO}_2$  at 90 °C in an automated acid-bath-preparation system using  $\text{H}_3\text{PO}_4$  and then measured on a Fisons-Prism isotope-ratio mass spectrometer (Fisons, Middlewich, Cheshire, UK). The analytical reproducibility of all  $\delta^{13}\text{C}$  measurements was  $<0.2\%$ .

### Stable-carbon-isotope balances

The increase in DIC concentrations in wells tapping the aquifer relative to the infiltrating river water from the River Alte Aare was assessed using stable-carbon-isotope balances, as described in Bolliger et al. (1999). As the groundwater flows through zones containing biodegraded organic material, it becomes enriched in DIC and the stable-carbon-isotope ratio of the incremental DIC,  $\delta^{13}\text{C}_{\text{inc}}$ , can reveal the nature of  $\text{DIC}_{\text{inc}}$  sources. This is expressed by the following two mass-balance equations for  $\text{DIC}_{\text{meas}}$  and  $\delta^{13}\text{C}_{\text{meas}} \times \text{DIC}_{\text{meas}}$  [Eqs. (2) and (3)]:

$$\text{DIC}_{\text{meas}} = \text{DIC}_{\text{river}} + \text{DIC}_{\text{inc}} \quad (2)$$

$$\delta^{13}\text{C}_{\text{meas}} \times \text{DIC}_{\text{meas}} = \delta^{13}\text{C}_{\text{river}} \times \text{DIC}_{\text{river}} + \delta^{13}\text{C}_{\text{inc}} \times \text{DIC}_{\text{inc}} \quad (3)$$

where

$\text{DIC}_{\text{meas}}$  measured DIC concentration in aquifer ( $\text{mol L}^{-1}$ )

$\text{DIC}_{\text{river}}$  measured DIC concentration in infiltrating river water ( $\text{mol L}^{-1}$ )

$\text{DIC}_{\text{inc}}$  DIC increase ( $\text{mol L}^{-1}$ )

$\delta^{13}\text{C}_{\text{meas}}$   $\delta^{13}\text{C}$  of  $\text{DIC}_{\text{meas}}$  (‰)

$\delta^{13}\text{C}_{\text{river}}$   $\delta^{13}\text{C}$  of  $\text{DIC}_{\text{river}}$  in infiltrating river water (‰)

$\delta^{13}\text{C}_{\text{inc}}$   $\delta^{13}\text{C}$  of  $\text{DIC}_{\text{inc}}$  (unknown) (‰)

The mass balance [Eq. (3)] has the form of a linear equation. Plotting the products of  $\delta^{13}\text{C}_{\text{meas}} \times \text{DIC}_{\text{meas}}$  versus  $\text{DIC}_{\text{inc}}$  allows the interpretation of the slope which is equal to the unknown  $\delta^{13}\text{C}_{\text{inc}}$ .

### Flow Modelling

In order to understand the impact of organic wastes on groundwater quality the local water-flow characteristics must be known. For this purpose we developed a quantitative 2-D flow model by using the commercial software package GMS, version 2.1 (Scientific Software Group). The program GMS is entirely based on the MODFLOW

code. The recently developed regional 2-D flow model (Geotechnisches Institut 1998) served as the basis for the local model. The regional model was refined and optimised by taking a smaller mesh size (25 m) and including this data set for piezometric heads, permeabilities and bank filtration along the River Alte Aare. Mass-transport calculations were performed with the MT3D module.

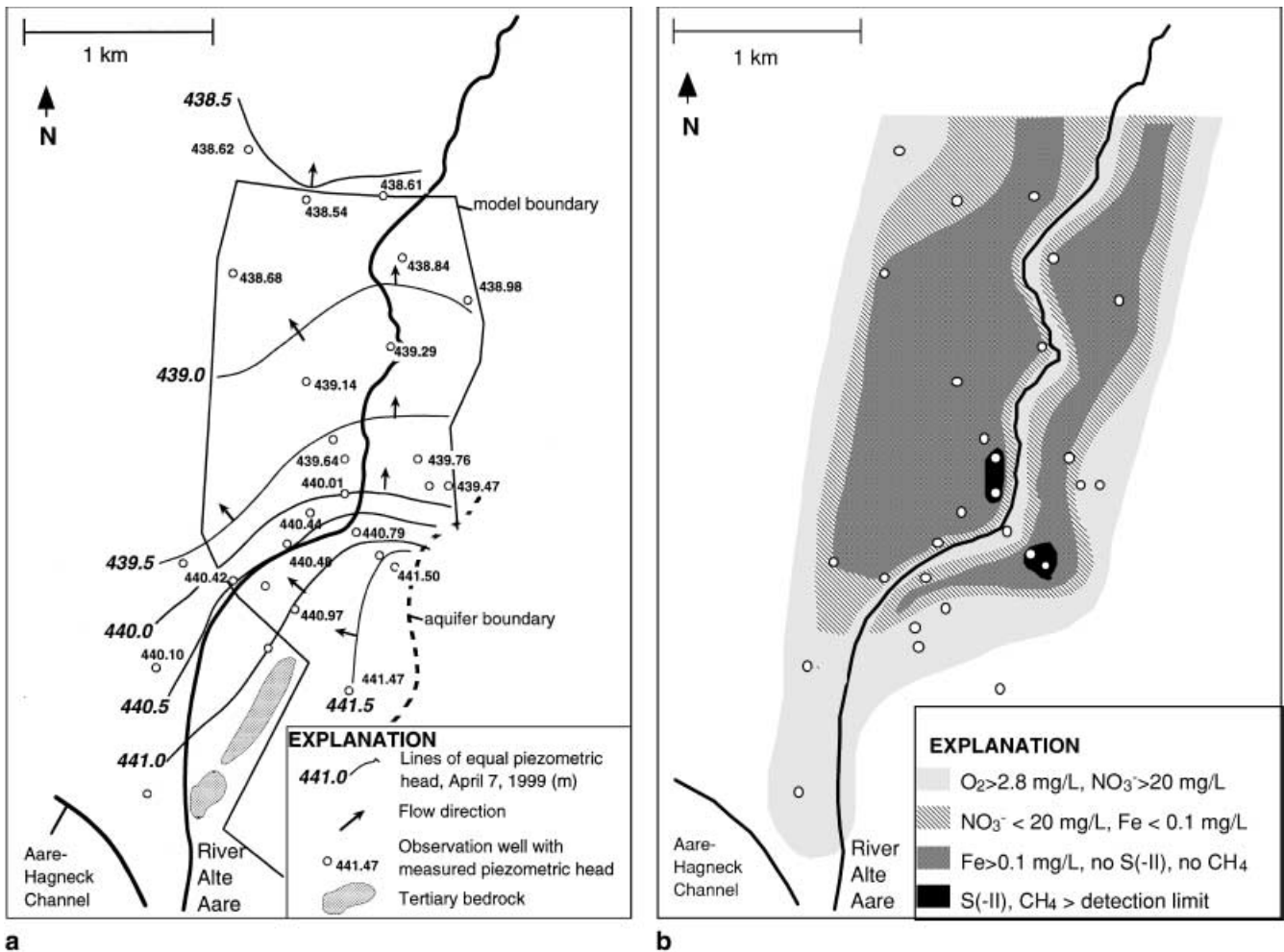
## Results and Discussion

### Groundwater Flow

From the piezometric head data it can be inferred that within the area of interest groundwater flow occurs from east to west and then turns to the north, subparallel to the River Alte Aare (Fig. 3a). Permeability data obtained from flowmeter measurements in 13 monitoring wells indicate K-values ranging from  $3 \times 10^{-4}$  to  $1.5 \times 10^{-2}$  m/s. Low transmissivities occur in the area of the Tertiary bedrock outcrop (Fig. 3a) whereas consistently higher transmissivities are noted downgradient thereof. Local specific infiltration rates, whose adequate assessment is crucial for the description of local hydraulic conditions, were obtained with the  $^{222}\text{radon}$  method (Table 1). The radon method can be applied for wells receiving river water with a mean residence time of from 0.5 to 15 d in the aquifer (Hoehn and von Gunten 1989). The average specific infiltration rate calculated from  $^{222}\text{radon}$  measurements in six wells was  $1.76 \pm 0.85 \text{ m}^3 \text{ m}^{-2} \text{ d}^{-1}$ . Multiplying this value by an estimated surface area of the river bed within the considered area ( $10,800 \text{ m}^2$ ), an overall infiltration rate of  $220 \pm 106 \text{ L/s}$  was obtained in April 1999. This compares with the former results of  $350 \text{ L/s}$  (Kellerhals and Haefeli 1988) obtained by flow measurements and  $140 \text{ L/s}$  (Geotechnisches Institut 1998) obtained from the regional-flow model.

A numerical flow model has been conceived using information on hydrogeology, measured hydraulic conductivities, measured river-water infiltration rates, and estimated recharge rates. This model has been refined using existing larger-scale flow models for the whole Bernese Seeland aquifer (Geotechnisches Institut 1998). The calibration procedure involved in a first step the modeling of a large area of about  $20 \text{ km}^2$  with the same hydraulic data and boundary conditions as the regional model and verification that the same flow conditions were obtained. In a further step the area was reduced to the size of interest and the hydraulic data collected were included to derive flow conditions in the area affected by SFA activities. In addition, available precipitation data were used to estimate aquifer recharge. The model boundaries to the east and west were defined by flow lines, the southeastern boundary constitutes the aquifer border, and the southern and northern boundaries are defined by equipotential lines (Figs. 1 and 3a).

The obtained fluxes in and out of the modelled area are shown in Table 2. The results indicate that for both periods of field measurements (October and April) nearly steady-state flow conditions can be inferred for the ar-



**Fig. 3a** Piezometric surface on 7 April 1999. **b** Redox conditions derived from parameters measured in groundwater samples on 7 April 1999

**Table 2** Water balance of flow model for the two situations in October 1998 and April 1999 (in  $L s^{-1}$ )

Type of in- and outflow	October 1998	April 1999
Inflow at southern boundary	5.5	2.1
Inflow at western boundary (along slope)	8.6	9.8
Precipitation	11.7	13.1
Bank infiltration	116.3	196.9
Total inflow	142.0	221.8
Outflow at northern boundary	141.7	223.3
Difference	0.2%	0.7%

ea of interest. The flow model underlines the importance of local river-water infiltration to the water balance. The modeled bank infiltration value obtained for April 1999 (197 L/s) is within the range of values obtained with the radon method on that selected sampling day  $220 \pm 106$  L/s. The aquifer is characterised by small-scale horizontal and vertical heterogeneities with regard to hydraulic properties. This is especially obvious in the area

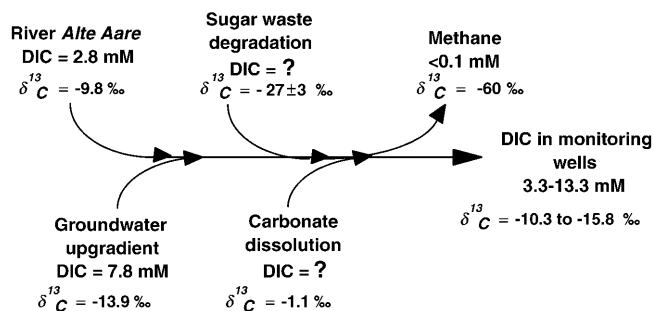
of river-bank filtration where the flowmeter (data not shown) and radon measurements (Table 1) indicate considerable variations in hydraulic conductivities with respect to specific infiltration rates. It is reasonable to assume that the infiltrating river water does not significantly mix with the groundwater in aquifer regions close to the river, and that therefore vertical heterogeneities in groundwater chemistry might occur. Since the groundwater chemistry data rely on sampling by pumping fully screened wells which extend down to the aquitard, groundwaters from different horizons are mixed during sampling. Despite these conditions, the application of a 2-D groundwater flow model and 2-D areal interpretations of groundwater chemistry are feasible because the focus of the study has been exclusively on the large-scale distribution of redox zones and fluxes of oxidants and reductants; therefore the effects of small-scale heterogeneity could be neglected.

### Groundwater Chemistry

Water from the two sampling periods October 1998 and April 1999 yielded a generally consistent picture of the distribution of redox-sensitive species in the groundwater (Tables 5 and 6). However, in April 1999, a general

trend towards higher O<sub>2</sub> saturations and lower concentrations of reduced species [Mn(II), Fe(II) and S(-II)] could be observed. This can be explained by more precipitation in spring 1999, leading to higher infiltration rates by the River Alte Aare. Upgradient of the zone influenced by wastes from the SFA, the groundwater composition is characterised by O<sub>2</sub> saturations >20%; low DOC, NH<sub>4</sub><sup>+</sup> and K<sup>+</sup> concentrations; and nitrate concentrations from 7–38 mg/L. In the zone influenced by SFA, there were lower O<sub>2</sub> and NO<sub>3</sub><sup>-</sup> concentrations and higher DOC, Mn(II), Fe(II), NH<sub>4</sub><sup>+</sup> and K<sup>+</sup> concentrations. These changes were accompanied by large increases in DIC (up to 20.4 mM) and also in Ca<sup>2+</sup> (up to 4 mM). In some of the wells, the groundwater chemistry is highly influenced by the infiltrating river water (Tables 5 and 6). This is reflected also by the <sup>222</sup>Rn measurements (Table 1). In two wells (V22, V24) located near a galvanic factory, high chloride concentrations have been observed.

The data on redox-sensitive species sampled in April 1999 were used to obtain a picture of the large-scale redox plume at the regional scale (Fig. 3b). The interpolated areas depicted in Fig. 3b were generated based on the criteria stated in the caption of the figure, and extrapolations were extended towards the borders of the map for lack of data there. South of well KB98/1, there is evidence of aerobic conditions (wells Aarfs01, KB98/1); however, sufficient wells are not available to resolve spatial heterogeneities in redox chemistry. Reduced conditions can be seen north of well KB98/1 on both sides of the River Alte Aare, with two “hot spots” on either side where S(-II) and methane were detected, thus indicating very low redox potentials. On the western side, the hot spot is located under the wastewater treatment plant from the SFA, and on the eastern side the hot spot coincides with a sugar-waste deposit dating from the 1940s and 1950s. Along both sides of the river are zones influenced by river infiltration where groundwater con-



**Fig. 4** Schematic illustration of processes influencing dissolved inorganic carbon (DIC) concentrations and its  $\delta^{13}\text{C}$  in groundwater

tains oxidants such as O<sub>2</sub> and NO<sub>3</sub><sup>-</sup>. This indicates the importance of river-water infiltration for the oxygenation of groundwater. During the sampling in October, these zones were less pronounced, indicating smaller contributions of oxygenated river-water infiltration then. The data obtained are similar to those obtained from other studies performed in the last decade (Halmes 1993; Hoffmeyer 1995; Geotechnisches Institut 1998). They confirm the general picture of a large-scale redox plume downstream of the SFA.

### Stable-Carbon Isotopes

The  $\delta^{13}\text{C}$  in DIC was measured in all monitoring wells sampled during the campaign in April 1999 (Table 6; see Appendix). The values range from -9.8‰ (river water) to -15.5‰ (V13) and -15.80‰ (VA). To interpret the variations in  $\delta^{13}\text{C}$  in DIC, a schematic model is proposed (Fig. 4). The  $\delta^{13}\text{C}$  in DIC of natural background groundwater has been measured in two wells (A1.1, Aarfs01) as -13.9‰, and in river water as -9.8‰. From literature data, it is inferred that DIC that has been mineralised from sugar residues or natural organic matter by non-

**Table 3** Summary of processes involved in sugar-residue mineralisation in a mixed aerobic and anaerobic aquifer and contributions to inorganic carbon and stable-carbon-isotope composition ( $\delta^{13}\text{C}$ ) of dissolved inorganic carbon. Species printed in *bold* are used for quantification of the process (processes 5 and 6 neglected). <CH<sub>2</sub>O> Sugar residues

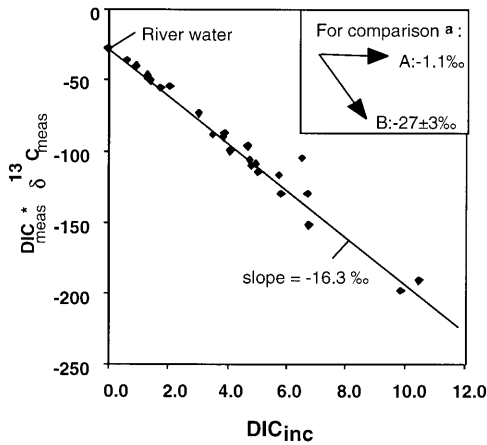
No.	Process	Contribution to inorganic C <sup>a</sup>	$\delta^{13}\text{C}$ of inorganic C (‰)
Mineralisation of sugar residues			
1	<b>O<sub>2</sub></b> +<CH <sub>2</sub> O>->CO <sub>2</sub> +H <sub>2</sub> O	+1	-27±3 <sup>b</sup>
2a	<b>NO<sub>3</sub><sup>-</sup></b> +5/4<CH <sub>2</sub> O>+H <sup>+</sup> ->5/4CO <sub>2</sub> +1/2N <sub>2</sub> +7/4H <sub>2</sub> O	+5/4	-27±3 <sup>b</sup>
2b	<b>NO<sub>3</sub><sup>-</sup></b> +2<CH <sub>2</sub> O>+2H <sup>+</sup> ->2CO <sub>2</sub> +NH <sub>4</sub> <sup>+</sup> +H <sub>2</sub> O	+2	-27±3 <sup>b</sup>
3	MnO <sub>2</sub> (s)+1/2<CH <sub>2</sub> O>+2H <sup>+</sup> ->1/2CO <sub>2</sub> + <b>Mn<sup>2+</sup></b> +3/2H <sub>2</sub> O	+1/2	-27±3 <sup>b</sup>
4	FeOOH(s)+1/4<CH <sub>2</sub> O>+2H <sup>+</sup> ->1/4CO <sub>2</sub> + <b>Fe<sup>2+</sup></b> +7/4H <sub>2</sub> O	+1/4	-27±3 <sup>b</sup>
5	SO <sub>4</sub> <sup>2-</sup> +2<CH <sub>2</sub> O>+2H <sup>+</sup> ->2CO <sub>2</sub> +H <sub>2</sub> S+2H <sub>2</sub> O	+2	-27±3 <sup>b</sup>
6	<CH <sub>2</sub> O>->1/2CO <sub>2</sub> +1/2CH <sub>4</sub>	+1/2	>+38 <sup>c</sup>
Chemical reactions			
7	CaCO <sub>3</sub> (s)+2H <sup>+</sup> ->CO <sub>2</sub> + <b>Ca<sup>2+</sup></b> +H <sub>2</sub> O	+1	-1.1 <sup>d</sup>
8	MgCO <sub>3</sub> (s)+2H <sup>+</sup> ->CO <sub>2</sub> + <b>Mg<sup>2+</sup></b> +H <sub>2</sub> O	+1	-1.1 <sup>d</sup>

<sup>a</sup> Moles per mole turnover of species in *bold*

<sup>b</sup> Corresponds to isotopic composition of sugar from sugar beets (Clark and Fritz 1997). The small amount of fractionation during mineralisation is neglected

<sup>c</sup> Methanogenic mineralisation is reported to enrich  $\delta^{13}\text{C}$  of DIC up to +38‰ (Grossman 1997)

<sup>d</sup> Measured  $\delta^{13}\text{C}$  of carbonate in a mixed sample from Bernese Seeland aquifer material (Bolliger et al. 1999)



<sup>a</sup> Vectors in the insert are for comparison of slopes to identify the source of DIC<sub>inc</sub>  
 A: DIC<sub>inc</sub> produced by carbonate dissolution  
 B: DIC<sub>inc</sub> produced by non-methanogenic mineralization

**Fig. 5** DIC<sub>meas</sub> × δ<sup>13</sup>C<sub>meas</sub> plotted versus DIC<sub>inc</sub> for all wells sampled in April 1999

methanogenic mineralisation has a δ<sup>13</sup>C in DIC of  $-27 \pm 3\text{‰}$  (Table 3; Clark and Fritz 1997). DIC originating from carbonate dissolution has δ<sup>13</sup>C in DIC of  $-1.1\text{‰}$ , and DIC from methanogenesis would yield positive δ<sup>13</sup>C of up to  $+38\text{‰}$  (Table 3). From the schematic model in Fig. 4 it can be inferred that the only process capable of lowering the δ<sup>13</sup>C in natural background groundwater is the non-methanogenic mineralisation of sugar residues or natural organic matter. A regional-scale map (Fig. 8) shows interpolated values of δ<sup>13</sup>C in DIC. This map illustrates that river-water infiltration is the main process driving δ<sup>13</sup>C towards high values and leading to a band of high ( $> -11\text{‰}$ ) values along both sides of the river. Two hot spots with δ<sup>13</sup>C values  $< -14\text{‰}$  can be located, coinciding approximately with the sites where the lowest redox conditions have been delineated (Fig. 3b).

Figure 5 is a graphical evaluation of the stable-carbon-isotope balance according to Eqs. (2) and (3). The regression line yields a slope of  $-16.3\text{‰}$ , which is an overall average of the unknown isotopic ratio of the produced DIC in the aquifer zone. When the influence of methanogenesis is excluded (only two wells had measurable methane concentrations), there were only two sources of DIC: non-methanogenic mineralisation ( $f_m$ ) and carbonate dissolution ( $f_c$ ). Based upon the distinct δ<sup>13</sup>C values of these processes, Eq. (4) is established (Bolliger et al. 1999).

$$f_m \times (-27 \pm 3\text{‰}) + f_c \times (-1.1\text{‰}) = -16.3\text{‰}. \quad (4)$$

Considering the fact that  $f_m + f_c = 1$ , Eq. (4) yields  $f_m = 0.59 \pm 0.07$ , signifying that for the average of all wells,  $59 \pm 7\%$  of the DIC increase originates from non-methanogenic mineralisation and  $41 \pm 7\%$  from carbonate dissolution.

### Leaching From Waste Deposits

The results, summarised in Table 7, show increased concentrations of DOC and COD in leachate with respect to groundwater concentrations. The observed concentrations are, however, 10–100 times lower compared to untreated sugar-factory waste waters (Hoffmann 1985). It is apparent that leachates from younger deposits display higher loading of organic carbon and dissolved salts. This is consistent with data from analyses of solids (data not shown) which indicate higher amounts of TOC and COD in younger deposits.

An estimate of mass losses from waste deposits was obtained as follows. The total surface of the waste deposits amounts to about 450,000 m<sup>2</sup>. The annual infiltration rate is about 300 L m<sup>-2</sup> year<sup>-1</sup>. Average DOC concentrations in leachate from waste samples (Table 7; see Appendix) are about 25 mg C/L. For the whole area, a leaching rate of 3.4 metric tons/year of carbon in the form of DOC can be calculated.

### Quantitative Analysis of Redox Processes

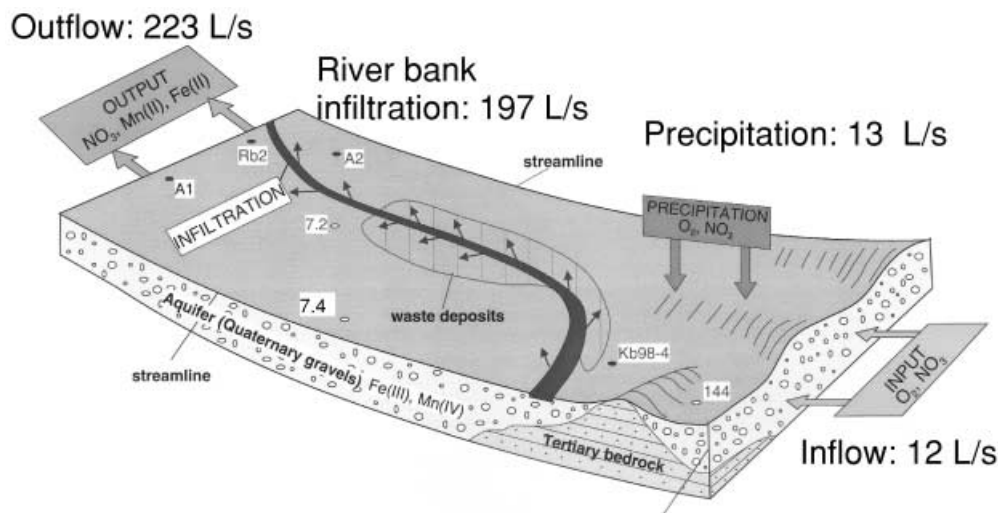
From the hydraulic and geochemical results presented in the previous sections, fluxes and degradation of organic carbon can be assessed. For this, a steady-state box model approach was used, in which biogeochemical processes related to carbon degradation are included. A box for the area of interest was defined based on the results of the groundwater analyses, on the availability of monitoring wells and on hydraulic constraints. This box is illustrated in Fig. 6. It is limited by the southern and southeastern borders of the aquifer upgradient and by the section along the wells A1, A2 and A3 downgradient of the waste deposits (Fig. 1). The steady-state box model for quantitative analysis of redox processes combines results from the flow model and from hydrochemical and C-isotope analysis. The approach includes the three steps described below.

In a first step the hydraulic conditions within the area of interest are elucidated with the aid of the groundwater flow model. The water flux into the box (Fig. 6) is governed by (1) the inflow of southern and southeastern groundwater, (2) river infiltration and (3) recharge by precipitation. According to the flow-model calculations, the outflow of water is roughly (within 2%) equal to the inflow for both periods investigated. Thus, steady-state flow conditions can be inferred.

In a second step, carbon fluxes in the inflows and outflow were estimated. From the hydrochemical results it can be deduced that the main dissolved C-fraction is DIC and only a small part exists as DOC. From the δ<sup>13</sup>C results it can be inferred that about 60% of DIC in groundwater downgradient of the waste deposits stems from organic-matter degradation and 40% from carbonate dissolution. Thus, by assessing the increase of DIC flux downgradient relative to upgradient, the amount of degraded organic C can be estimated. The mass-balance calculations are done by taking into account the results of the flow model for the autumn and spring situations.



**Fig. 6** Schematic representation of the steady-state box model approach, with estimated water fluxes for 7 April 1999



The increase in the DIC flux is then calculated by a simple mass balance:

$$\Delta\text{DIC}_{\text{box}} Q_{\text{out}} = Q_{\text{out}}[\text{DIC}]_{\text{out}} - (Q_{\text{i}}[\text{DIC}]_{\text{i}} + Q_{\text{ii}}[\text{DIC}]_{\text{ii}} + Q_{\text{iii}}[\text{DIC}]_{\text{iii}})$$

and

$$R_{\text{Corg}} = 0.6 \times \Delta\text{DIC}_{\text{box}} \times Q_{\text{out}} \text{ (mol/s) where } Q_{\text{out}} = Q_{\text{i}} + Q_{\text{ii}} + Q_{\text{iii}}$$

where  $\Delta\text{DIC}_{\text{box}}$  is the amount of DIC increase in the considered area (mmol/L);  $Q$  and  $[\text{DIC}]$  are the water flow (L/s) and the DIC concentrations (mol/L) in the outflow; the different respective inflows are i (from upgradient), ii (river infiltration), and iii (recharge); and  $R_{\text{Corg}}$  (mmol/s) is the overall rate of organic-carbon degradation within the modelled area.

In a third step, the relative contribution of the different redox processes to carbon degradation was estimated. The transformation of organic carbon via interaction with electron acceptors leads to an increase in DIC (Table 3). The hydrochemical results indicate that  $\text{O}_2$ , nitrate, manganese(IV) and iron(III) are reduced as a result of microbial activity. The possible reduction of  $\text{SO}_4^{2-}$  and production of methane are neglected in this consideration. The mass balance for  $\text{O}_2$  and  $\text{NO}_3^-$  can be assessed analogously to the one described for DIC. For Mn and Fe, however, this simple approach is not straightforward, since reduced solid phases in the aquifer, such as  $\text{FeS}$  or  $\text{FeCO}_3$ , cannot be quantified easily. We have roughly estimated the rate of Mn and Fe reduction by subtracting the  $\text{O}_2$  and  $\text{NO}_3^-$  transformation rate from the overall degradation rate  $R_{\text{Corg}}$ . The results are summarised in Table 4 for both the October and April situations.

The mass-balance calculations show the following results:

1. The overall degradation rate in the modelled area is 115 mmol/s (Table 4). This corresponds to 43 metric tons of carbon per year.
2. The degradation rate is similar in both periods investigated which suggests that the higher-flow conditions

**Table 4** Calculated overall transformation rates in modelled area (in mmol/s)

Transformation rates	October 1998	April 1999	Comments
$R_{\text{orgC}}$	-116.3	-113.0	From DIC balance
$R_{\text{O}_2}$	-38.0	-66.9	From $\text{O}_2$ balance
$R_{\text{NO}_3^-}$	-27.7	-2.8	From $\text{NO}_3^-$ balance
$R_{\text{Fe+Mn}}$	-50.7	-42.9	$R_{\text{Fe+Mn}} = R_{\text{orgC}} - R_{\text{O}_2} - R_{\text{NO}_3^-}$

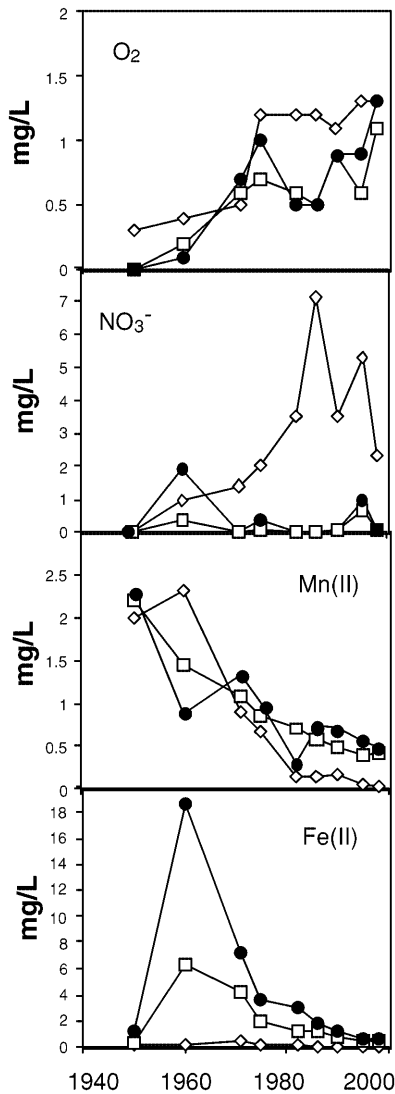
in spring compensate for the lower redox-transformation rates.

3. The flux of degraded carbon is higher than the presumed total flux of dissolved organic carbon leached from the waste sites by a factor of about 10 (3.4 metric tons of carbon per year). The latter estimate is based on results of the study of leachate from the waste deposits.
4. The average flux obtained for degraded organic carbon in the area affected by SFA activities (115 mmol/s) is a factor of 2 to 3 higher than the flux of consumed  $\text{O}_2$  (38–67 mmol/s; Table 4). This is in qualitative agreement with the observed anoxic conditions in the area.

The most probable explanation for these findings lies in the nature of the former practice of wastewater disposal. According to the study of Bruderer et al. (1971), about 650 t of degradable organic carbon per year (650 mmol/s) was disposed as wastewater in settling ponds within the period 1940–1964. Significant quantities of this organic carbon still seem to be present in the aquifer.

#### Temporal Evolution of Redox Conditions

Since 1950 a monitoring programme of the groundwater quality in the Bernese Seeland has been performed by the water-supply company of the Seeland (Wasserversorgung Seeland 1999). Analysis of the hydrochemi-



**Fig. 7** Temporal evolution of concentrations of O<sub>2</sub>, NO<sub>3</sub><sup>-</sup>, Mn(II) and Fe(II) in wells A1 (open diamonds), A2 (open squares) and A3 (solid circles) between 1950 and 1999 downgradient of the sugar factory at Aarberg, measured in November or December of each year. (Data from Wasserversorgung Seeland 1999)

cal data for observation wells A1, A2 and A3, located downgradient of the waste deposits, indicates a general trend to less anoxic conditions with time. As illustrated in Fig. 7, an increase in O<sub>2</sub> and NO<sub>3</sub><sup>-</sup> and a decrease in dissolved Fe and Mn are noted between 1960 and 1980. The trend for the most recent period (1980–1998) is less obvious, suggesting more constant redox conditions. These findings are consistent with the historical reconstruction of wastewater data (Fig. 2), suggesting that the maximum DOC load to the aquifer extended to the beginning of the 1960s and then decreased. Mean groundwater-flow velocities in the region studied can be inferred from the groundwater model used in this study and are in the order of 500 m/year. Thus, it would take 5 years for a dissolved plume to migrate towards monitoring wells A1, A2 and A3, which form the downgradi-

ent border of this steady-state box model. Presumably the slow degradation of the large pool of “old” sugar-waste residues in the subsurface is the main reason for the prolonged presence of reduced conditions in the study area.

## Conclusions

A combined study including hydraulic, leachate, hydrochemical and stable-carbon-isotope analyses enabled the characterisation of a large-scale redox plume that developed from former liquid- and solid-waste disposal practices of the sugar factory of Aarberg. The efforts of this study concentrated on the source area, i.e. the zone containing the sugar-waste deposits. Thus, the extension of the reduced zones downgradient of the study area beyond a line connecting wells A3, RB2 and A1.1 has not been determined in this work. Also, due to the limited number of monitoring wells the lateral boundaries in some areas of the redox plume could not be precisely defined.

This study allows a better quantitative understanding of processes involved in the redox-plume generation next to the presumed source zones. It is concluded that at present an excess input of molecular oxygen to the groundwater has been re-established compared to DOC. Monitoring data from 1960 to the present, downstream of the sugar-waste deposits, suggest a slow improvement in water quality, which shows a tendency to shifting from anoxic to suboxic conditions. Because of the large amount of previously released sugar-waste materials and their metabolites that are still present in the subsurface, the re-establishment of oxic conditions may take many decades.

Measures to enhance this process in the study area are (1) to further minimise leachate generation from waste-deposits, and (2) to increase river-water infiltration rates. Studies to manage the latter by preventing river-bed sealing during dry-weather periods of low-river discharge are currently envisaged.

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## Appendix

The additional Fig. 8 shows the interpolated pattern of  $\delta^{13}\text{C}$  values measured in DIC on 7 April 1999. Tables 5 and 6 show the results of groundwater and river analyses from October 1998 and from April 1999 respectively. Table 7 shows the results of leachate experiments with soil samples from waste deposits.

**Fig. 8** Map showing interpolated pattern of  $\delta^{13}\text{C}$  values measured in DIC on 7 April 1999

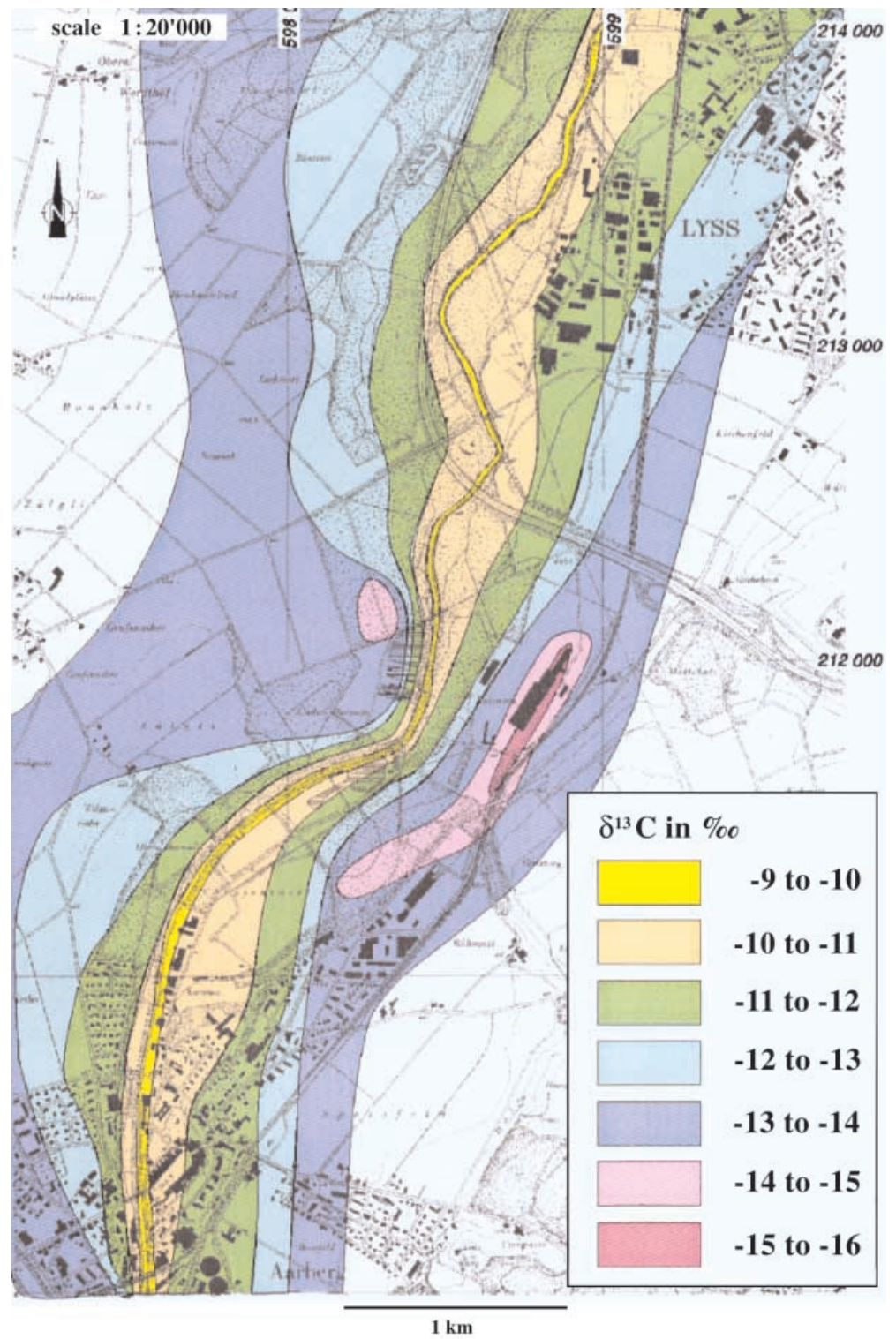


Table 5 Results of groundwater and river analyses from October 1998. *n.m.*, Not measured

Well (River)	Temp. (°C)	pH	O <sub>2</sub> (mg/L)	Cl <sup>-</sup> (mg/L)	SO <sub>4</sub> <sup>-</sup> (mg/L)	NO <sub>3</sub> <sup>-</sup> (mg/L)	NH <sub>4</sub> <sup>+</sup> (mg/L)	K <sup>+</sup> (mg/L)	Ca <sup>2+</sup> (mg/L)	Na <sup>+</sup> (mg/L)	Mg <sup>2+</sup> (mg/L)	Mn(II) (mg/L)	Fe(II) (mg/L)	DOC (mg/L)	S(-II) (µm)	CH <sub>4</sub> (µm)	Alk. (mEq/L)	DIC (mM)	δ <sup>13</sup> C (‰)	
Kb98/2	14.3	7.22	1.68	9.2	30	10.6	n.m.	2.9	77	5.6	n.m.	0.04	0.03	0.7	n.m.	n.m.	3.9	4.5	n.m.	
Kb98/3	15.3	7.18	2.28	5.2	29	3.46	1.7	4.5	75	6.4	n.m.	0.24	0.59	1.5	n.m.	n.m.	4	4.6	n.m.	
Kb98/4	12.7	7	4.63	14.8	22	28.4	0.1	4.8	129	9.9	n.m.	0.21	0.18	0.9	n.m.	n.m.	6.3	7.9	n.m.	
PmD1	12.5	6.95	0.10	28	46	5.8	n.m.	9.2	144	11	n.m.	0.18	0.02	2.1	n.m.	n.m.	8	10.2	n.m.	
Möri	12.3	6.94	0.23	8.8	53	7.2	n.m.	11.4	123	8.2	n.m.	0.01	0	1.6	n.m.	n.m.	6.2	8.0	n.m.	
Rb1	15.7	7.4	0.75	n.m.	34	0	n.m.	1.8	64	3.1	n.m.	0.19	0.2	0.5	n.m.	n.m.	2.9	3.2	n.m.	
Rb2	12	7.39	0.57	n.m.	31	1.8	n.m.	1.7	67	3.3	n.m.	0	0	0	n.m.	n.m.	3.3	3.6	n.m.	
7.1	11	7.03	0.77	n.m.	35	4.2	n.m.	2.3	113	9.7	n.m.	0.13	0.05	0.6	n.m.	n.m.	6.4	7.9	n.m.	
7.2	11.3	7.16	1.27	5.4	44	4.5	n.m.	9.6	108	4.2	n.m.	0.14	0.03	0.9	n.m.	n.m.	5.5	6.5	n.m.	
7.4	11.2	7.16	1.45	n.m.	36	3	n.m.	4.1	95	5.8	n.m.	0.26	0.15	0.6	n.m.	n.m.	4.7	5.5	n.m.	
7.5	13.6	7.39	2.92	n.m.	34	6.9	n.m.	2.2	72	3.2	n.m.	0	0.01	0	n.m.	n.m.	3.2	3.5	n.m.	
144	11.7	7.06	5.04	n.m.	30	15.1	n.m.	5.2	106	9.9	n.m.	0	0.02	0	n.m.	n.m.	5.4	6.6	n.m.	
V10	11.5	6.92	1.26	13	29	25.2	0	7.5	128	8.7	12.7	0.14	0.87	2.2	n.m.	n.m.	10.9	8.7	n.m.	
V12	10.7	6.85	0.63	26	116	0	0.67	18.4	163	15	46	0.54	3.6	6.9	n.m.	n.m.	6.69	14.9	n.m.	
V14	12.4	7.01	0.52	10.2	23	10.6	0.94	3.6	103	6.3	9.3	0	0	0.86	n.m.	n.m.	5.39	6.7	n.m.	
ZRA2	13.8	6.62	0.63	77	21	8.86	0.08	130	137	57	16.5	0.61	5.3	9.2	n.m.	n.m.	12.9	20.4	n.m.	
V25	11.2	7.12	0.52	10.2	33	2.7	0.39	13.3	120	7.4	8.7	2.1	0.62	1.41	n.m.	n.m.	7.8	7.8	n.m.	
ZRA1	15.1	7.05	0.67	2.7	30	0	0.06	2.3	77	2.8	4.7	0.47	0.57	1.62	n.m.	n.m.	3.74	4.5	n.m.	
V22	14.2	6.77	0.63	22	60	6.11	0.04	24	143	11.5	26	0.13	0	4	n.m.	n.m.	8.74	12.3	n.m.	
A2	11.2	7.61	0.42	6.1	33	0	0.05	2.5	72	4.9	5.6	0.44	3.5	1.56	n.m.	n.m.	3.66	3.9	n.m.	
A1.1	10.8	7.07	2.20	16.5	34	27.5	0	2.9	138	6.3	10	0	0	0.83	n.m.	n.m.	6.72	8.2	n.m.	
A1	11.1	7.52	0.31	8.9	37	0	0	5.8	94	6.7	7	0	4.1	1.61	n.m.	n.m.	4.83	5.2	n.m.	
River	12.7	7.95	5.66	4.7	36	4.7	n.m.	2	62	4.1	n.m.	0.15	0.04	1.6	n.m.	n.m.	2.7	2.8	n.m.	
Alte																				
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Table 6 Results of groundwater and river analyses from April 1999. n.m. Not measured

Well (River)	Temp. (°C)	pH	O <sub>2</sub> (mg/L)	Cl <sup>-</sup> (mg/L)	SO <sub>4</sub> <sup>-</sup> (mg/L)	NO <sub>3</sub> <sup>-</sup> (mg/L)	NH <sub>4</sub> <sup>+</sup> (mg/L)	K <sup>+</sup> (mg/L)	Ca <sup>2+</sup> (mg/L)	Na <sup>+</sup> (mg/L)	Mg <sup>2+</sup> (mg/L)	Mn(II) (mg/L)	Fe(II) (mg/L)	DOC (mg/L)	S(-II) (μM)	CH <sub>4</sub> (μM)	Alk. (mEq/L)	DIC (mM)	δ <sup>13</sup> C (‰)	
Kb98/1	8.3	7.13	8.1	9.53	51.2	37.5	0.01	2.21	108	6.24	11.3	0.059	0.005	4.0	<1	0	7.8	9.4	-11.1	
Kb98/2	8.7	7.27	3.76	7.4	31.1	12.9	0.03	1.93	62.7	7.91	5.98	0.056	0.013	0.8	<1	0	4.3	4.9	-11.1	
Kb98/3	6.4	7.26	5.9	5.57	30.7	8	0	1.26	51.3	4.11	4.38	0.051	0.011	0.7	<1	0	3.6	4.2	-11.2	
Kb98/4	11.4	7.2	7.11	14.1	20.3	26.4	0.01	4.5	94.7	8.56	7.49	0	0	0.0	<1	0	6.0	7.0	-14.3	
V10	9.8	7.06	2.76	12.9	31.6	45.6	0.02	7.65	122	8.54	10.7	0	0	1.3	<1	0	6.4	7.9	-14.5	
V12	9	7.02	0.22	16.3	75.2	31.1	0.32	17.1	112	10.4	47.2	0.63	0.57	3.9	n.m.	1	10.6	13.3	-14.3	
V13	8.9	6.83	0.64	12.4	21.6	13.5	5.45	6.26	145	8.14	10.2	4.07	0.11	3.7	3	20	9.1	12.7	-15.5	
V14	10.7	7.08	0.29	10.5	25.7	30.6	0.11	4.61	90.3	7.44	9.03	0	0.017	1.4	<1	0	6.3	7.6	-13.9	
VA	n.m.	6.97	8.16	83.2	22.1	45.1	0.36	11.2	103	4.59	7.16	0	0.098	0.6	<1	0	7.0	9.6	-15.8	
V24	n.m.	7.09	8	6.6	32.1	32	0	6.64	126	7.81	9.25	0.021	0.021	1.7	<1	0	5.3	6.7	-13.3	
V25	9.5	7.04	1.15	7.95	33.9	12	0.04	2.98	241	18.9	12.6	0.015	0.059	1.3	<1	0	7.0	8.7	-14.9	
Rb1	10	7.61	0.19	10.2	40	0.73	0.02	17.1	85.6	6.23	7.55	0	0.051	0.0	<1	0	3.3	3.5	-10.3	
Rb2	0.9	7.43	2.11	8.5	30	5.49	0.01	1.2	53.4	5.46	4.76	0.053	0.052	0.0	<1	0	3.7	4.2	-11.6	
ZRA1	12.2	7.06	0.14	7.75	36.6	0	1.08	26.6	97.8	25.7	25.6	2.07	1.26	1.5	<1	2	6.3	7.6	-14.3	
ZRA2	11.8	7.02	0.2	7.65	10.3	0	1.45	9.88	87.4	6.32	6.94	3.6	1.06	2.6	14	110	7.7	9.6	-13.5	
7.1	10.7	7.09	0.82	13.8	25.5	8.31	0	2.04	76.3	6.78	9.89	0.07	0.026	0.5	<1	0	7.1	8.6	-13.6	
7.2	8.8	6.96	1.28	5.9	33.9	19.4	0.02	8.77	82.5	3.36	8.69	0	0.071	0.9	<1	0	5.8	7.5	-12.7	
7.4	11.2	7.13	0.72	11.5	35.8	6.05	0.01	5.5	78.5	6.03	7.96	0.031	0.24	0.7	<1	0	5.7	6.7	-12.9	
7.5	11.5	7.34	5.67	8.9	33.2	14.7	0	1.68	61.5	5.23	5.18	0	0.009	0.0	<1	0	3.8	4.2	-11.8	
A1	10.2	7.18	1	7.8	32.8	7.55	0.02	5.23	68.4	4.45	5.64	0	0.008	0.8	<1	0	5.0	5.9	-12.4	
A1.1	9.1	7.06	3.96	11.3	26.9	31.4	0.02	1.15	98.1	4.23	7.45	0	0	0.6	<1	0	6.3	7.8	-13.9	
A2	11.5	7.46	0.14	7.1	34.1	0	0.03	1.06	43.5	4.02	3.67	0	0.47	0.7	<1	0	3.5	3.8	-10.6	
A3	11	7.45	0.23	6.6	33.2	0	0.31	6.73	55.5	4.92	4.78	0	0.59	0.9	n.m.	3	4.2	4.6	-12.0	
AarFs01	12.5	7.12	2.58	20.6	31.3	11.2	0	8.86	73.7	11.2	6.8	0	0	0.7	<1	0	5.4	6.4	-13.8	
River	8.2	8.05	10.74	3.8	28.9	4.19	0.02	1.03	32.1	3.03	3.11	0	0.007	0.0	<1	0	2.8	2.9	-9.8	
Alte																				
Aare																				

**Table 7** Results of leachate experiments with soil samples from waste deposits

Sample no.	Waste deposit	Approx. year of deposit	K (mg/L)	Na (mg/L)	SO <sub>4</sub> (mg/L)	DOC (mg C/L)	COD (mg O <sub>2</sub> /L)	BOD <sub>5</sub> (mg O <sub>2</sub> /L)
Leachate								
E(AB1A)	AB1A	ca. 1915	5.5	0.7	<5	12.6	85.1	10.8
E(AB4)	AB4	ca. 1930	3.4	0.8	6.8	6.5	74	<5
E(AB6)	AB6	ca. 1950	6.2	0.6	7.2	12.5	73	
E(AB12)	AB12	ca. 1960	31.3	5.3	8.1	25.0	127	78
E1(D4)	D4	ca. 1970	31.9	8.3	6.4	24.8	338	
E2(D4)	D4	ca. 1970	37.4	10.0	12.3	21.9	316	
E(D1)	D1	ca. 1975	51.2	24.4	<5	29.3	191	87
E1(D2)	D2	ca. 1985	18.2	1.1	22.8	11.9	83	6
E2(D2)	D2	ca. 1985	17.2	1.2	<5	23.3	81	
SW(D2)	D2	ca. 1985	25.0	6	141.0	24.0	63.0	
SW(AB11)	AB11	ca. 1960	3.1	3.9	29.0	6.5	19.0	
Wastewater								
Untreated <sup>a</sup>		Since 1989	230–730		30–65	3,000–7,800	10,000–20,000	
Treated <sup>b</sup>		1997	257			10	50	5
Untreated historical <sup>c</sup>		1964				150–380		600–900

<sup>a</sup>At the entry of the wastewater treatment plant<sup>b</sup>At the exit of the wastewater treatment plant<sup>c</sup>Based on documents from the SFA

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