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VULNERABILITY OF A KARST SPRING TO WASTEWATER INFILTRATION (GALLUSQUELLE, SOUTHWEST GERMANY)

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ABSTRACT

Karst aquifers represent important drinking water resources worldwide. However, they exhibit a high vulnerability against contamination with chemicals and faecal bacteria due to rapid uptake and transport of pollutants via highly permeable solution conduits. In this study a spring sustained by such an aquifer, the “Gallusquelle” (Swabian Alb, Southwest Germany) was investigated with respect to wastewater-borne contamination. A rain spillway basin serving as a reservoir for excess of combined wastewater is located within the catchment of the spring and occasionally overflows after heavy rain events. Water volumes in the range of several thousand cubic metres are thus released into a dry valley. This study follows and quantifies the consequences of these overflows for the raw water quality at the karst spring. The main objective was to verify the connection between the infiltration of wastewater in the catchment and the decreasing water quality at the spring which had previously been suggested based on a tracer experiment. Therefore, time series of both microbiological as well as geochemical parameters of the spring water were analysed, suggesting that the wastewater from the rain spillway basin represents an important contamination source of the spring water.

Karstgrundwasserleiter haben als Trinkwasserressourcen weltweit eine große Bedeutung. Sie sind jedoch sehr anfällig für Verunreinigungen durch Chemikalien und Fäkalbakterien, da über das Röhrensystem Schad- und Schmutzstoffe sehr schnell aufgenommen und transportiert werden können. In der vorliegenden Studie wurde die Gallusquelle (Schwäbische Alb, Südwestdeutschland), die solch ein System entwässert, im Hinblick auf abwasserbedingte Verunreinigungen untersucht. Ein Regenüberlaufbecken, das überschüssiges Mischabwasser aufnehmen soll, befindet sich im Einzugsgebiet der Quelle und läuft nach Starkregenereignissen gelegentlich über. Wassermengen im Bereich von mehreren Tausend Kubikmetern werden dadurch in ein Trockental abgegeben. Diese Studie dokumentiert und quantifiziert die Auswirkungen solcher Überlaufereignisse auf die Rohwasserqualität der Karstquelle. Hauptziel war es, die Verbindung zwischen der Versickerung von Abwasser im Einzugsgebiet und der Verschlechterung der Wasserqualität an der Quelle zu verifizieren, welche man bereits aufgrund der Ergebnisse eines Markierversuchs vermutete. Hierzu wurden Zeitreihen sowohl mikrobiologischer als auch geochemischer Parameter des Quellwassers analysiert. Es sollte gezeigt werden, dass die Verschmutzung des Quellwassers im Wesentlichen durch das Abwasser aus dem Regenüberlaufbecken verursacht wurde.

1. INTRODUCTION

Groundwater from karst aquifers provides a considerable drinking water resource worldwide. Besides their heterogeneous hydrogeological properties these aquifers exhibit a special vulnerability against anthropogenic contamination due to their potential of fast uptake of recharge water and low natural attenuation capacity. The Gallusquelle spring as the main object of this study is sustained by a karst catchment located on the plateau of the western Swabian Alb, Germany and feeds into the Lauchert river (Fig. 1). Topographically, the terrain is located approximately between 600 m.a.s.l. at the spring level and 900 m.a.s.l. at the outermost boundary of the catchment (Fig. 2). The spring water is used for drinking water supply by a public water supply association. The raw water is abstracted and treated by a waterworks located in the vicinity of the spring. Problems with temporarily bad raw water quality documented by turbidity measurements and sporadic microbiological analyses have been occurring for many years (Sauter, 1992 and pers. communication by waterworks). A rain spillway basin located within the catchment at a distance of approximately 9 km from the spring is supposed to be the most important source for contamination (Fig. 1). As a consequence of heavy rain events, this artificial reservoir for combined wastewater occasionally overflows. During an average overflow event the basin, which has a volume of 6,500 m^3, releases wastewater volumes in the order of several thousand cubic metres into a dry valley (Harthauser Tal) in the karst region where infiltration is quite rapid. In this study the influence of wastewater inflow into the groundwater system was assessed by analysing the spring water for the number of faecal indicator bacteria, turbidity, and for concentrations of organic matter, major ions and organophosphates, that are typical for wastewater.

The dominating land cover around the rain spillway basin is forest, especially in the upper course of the dry valley, whereas the area at a distance of 3 to 6 km to the spring is mainly used for agriculture, both for grassland and grain growing. Grazing plays a minor role. The aquifer system of the Gallusquelle is formed in the Upper Jurassic carbonate rocks of the Swabian Alb, where the calcareous marls and the limestones of the Oxford 1 and 2 form mainly the aquifer base. The main aquifer, which is unconfined, is represented by the 80 to 170 m thick layer of micritic limestones of the Kimmeridgian 2/3 (ki2/3) and in the southeastern section by the marls of the Kimmeridgian 1. Near the northwestern boundary also the Oxford 2 layer contributes to the aquifer. At the catchment surface, the ki 2/3 and - in former submarine depressions and within the Hohenzollerngraben zone - the Tithonian, the youngest jurassic formation, crop out. The Hohenzollerngraben is a major fault zone within the catchment with direction northwest-southeast and throws between 70 and 100 m. Karstification within the fault zone is similarly developed as in the surrounding area, but storage is assumed to be higher due to the lowering of the ki2/3 layer. The aforementioned Harthauser Tal dry valley in its upper course runs within the boundaries of the graben structure (Fig. 1). The elevation of the water table decreases significantly in a distance of approximately 6 km from the spring (Fig. 2) Southeast of this inclination a highly karstified zone was documented, which is probably due to lithological heterogeneity (Sauter, 1992).

Flow velocities in the conduit system were determined in various tracer experiments with injection into sinkholes distributed over the whole area (Gwinner, 1973, Merkel, 1991, Sauter, 1992, Birk et al., 2005). Merkel (1991) for instance reported a high maximum tracer velocity of 160 m h^{-1} for an experiment with injection near the town of Bitz (Fig. 1). Where surface runoff can easily reach the conduit system, also biological and chemical contaminants can be transported quickly. Thus, even at distances of many kilometres from the spring, groundwater residence times may be below the limit (50 days in Germany, 60 days in Austria) that classifies an area as groundwater protection zone. From a practical point of view, however, it is not feasible to assign protection zones covering tens of square kilometres.

2. MATERIALS AND METHODS

Changes in discharge and water quality of the Gallusquelle were monitored over five months focussing on responses to heavy rain events and overflows of the rain spillway basin Bitz (RSpB). To this end, a multi-parameter probe system was used for online measurement of water level, electrical conductivity, turbidity, and water temperature. Water level measurements were converted to discharge values using a stage-discharge relationship developed by the local water authority. In addition to the automatically recorded data, pH was measured with a hand held meter, and alkalinity was determined by pH titration (methyl red method) on site. Precipitation and air temperature data were provided by a private meteorological station located in Bitz.

Water samples were taken at the Gallusquelle for analysis of microbiological parameters, content of total and dissolved organic carbon (TOC and DOC), and of major ions, respectively. Samples were collected once a week during dry periods; after the overflow events the sampling frequency was increased up to four samples per day. Additional water samples were collected occasionally at the rain spillway basin. The samples for DOC/TOC and ion analysis were collected and stored at 4 °C until they were analysed. DOC and TOC were determined with a high temperature TOC Analyser (Elementar Analysensystem GmbH). For the ion analysis samples were filtered and then measured with a standard ion chromatograph (DX-120, Dionex). Samples for analysis of organophosphates (only last overflow event) were collected in 2 l-glass bottles (Schott). Within two days they were shipped to the laboratory, where the organophosphates were extracted within 24 h using a liquidliquid extraction method. The samples were quantified by using large volume injection-GC-MS. The analytical method is described by Andresen et al. (2004) and Meyer and Bester (2004).

As a measure for water quality, four different microbiological parameters were quantified:

a. Total number of bacteria that form colonies at 20°C
b. Total number of bacteria that form colonies at 36°C
c. Number of coliform bacteria
d. Number of *Escherichia coli*.

For the determination of the parameters "a" and "b" the plate count method according to German Drinking Water Ordinance was applied. Coliform bacteria and *E. coli* (parameters "c" and "d") were detected and enumerated with Chromocult® Coliform Agar (CCA).

The proportion of overflow water in the spring water was estimated using a mass balance approach suggested by Dreiss (1989). It is based on two flow components, here assumed as

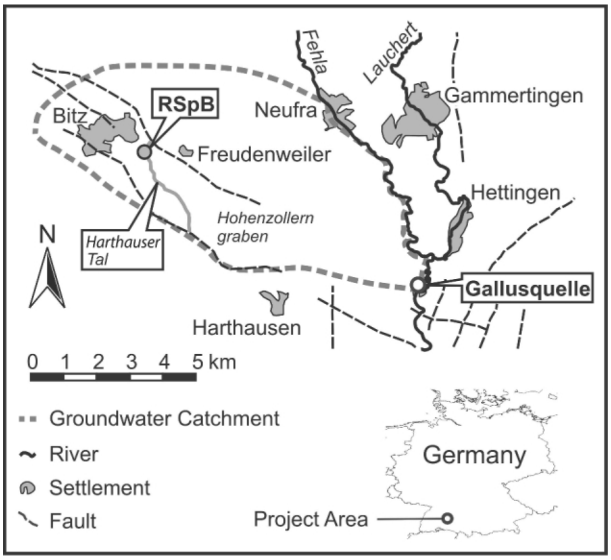

FIGURE 1: Location of the study area and of the rain spillway basin (RSpB) within the Gallusquelle spring catchment (modified from Birk et al., 2005).

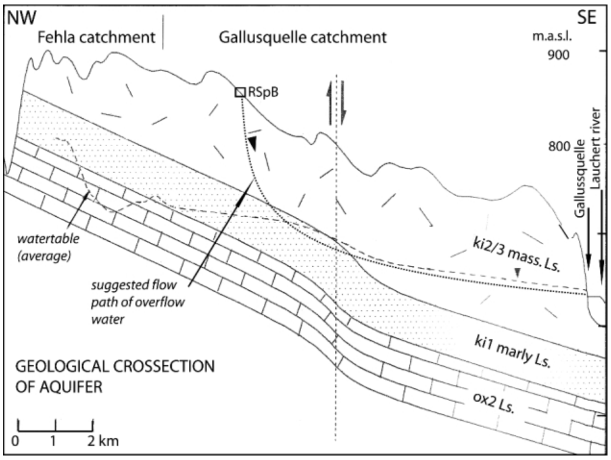

Figure 2: Geological structure of the Gallusquelle aquifer and topographical heights. RSpB = Rain spillway basin; ki2/3 mass. Ls. = Kimmeridge 2/3 massive limestones, ki1 marly Ls. = Kimmeridge 1 marly limestones, ox2 Ls. = Oxford2 limestones (aquifer base); assumed flexure in approx. 6 km NW of the Gallusquelle due to subsurface faulting (modified from Sauter, 1992).

the background flow Q_b and the flow from the rain spillway basin Q_{ofl}, thus neglecting the remaining event flow:

$$Q_s C_s = Q_b C_b + Q_{ofl} C_{ofl} \qquad (1)$$

where Q_s = spring discharge [L^3T^{-1}], C_s = measured concentration of compound during event response [ML^{-3}], C_b = background concentration of compound [ML^{-3}], C_{ofl} = measured concentration of compound in overflow water [ML^{-3}]. With $Q_s = Q_b + Q_{ofl}$ equation (1) can be solved yielding

$$Q_{ofl} = Q_s \frac{C_s - C_b}{C_{ofl}} \qquad (2)$$

The total volume of overflow water for one event is then estimated by

$$V_{ofl} = \int_0^\infty Q_{ofl}(t)dt \qquad (3)$$

where V_{ofl} = total volume of overflow water [L^3].

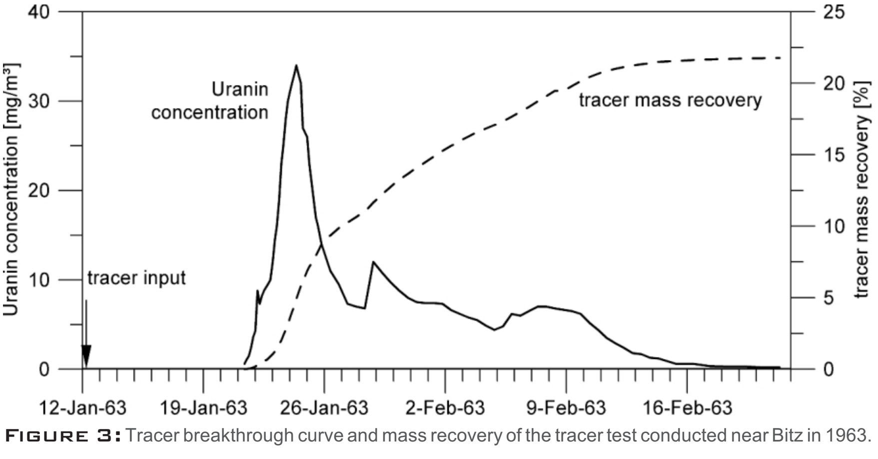

Figure 3: Tracer breakthrough curve and mass recovery of the tracer test conducted near Bitz in 1963.

3. Results and Discussion

In addition to the data obtained from the investigations described in the previous section, results from a tracer test conducted in the dry valley (Harthauser Tal) that drains the overflow of the rain spillway basin were available. The results from this artificial groundwater tracing experiment are presented in the following section, as they demonstrate that there is a fast transport pathway between the location of the rain spillway basin and the karst spring. Next, results from the investigations conducted in this study are presented.

3.1 Artificial Groundwater Tracing

Water contamination at the Gallusquelle was observed already in the 1960s. By that time the Harthauser Tal valley in its upper course was used as release trench for wastewater of the community of Bitz. To find out whether a hydraulic connection was existent between the infiltration area in the valley and the Gallusquelle, a tracer test was carried out by the Geological State Office of Baden-Württemberg in 1963 (Kiberler, 1963). Uranin (10 kg) was injected into a creek that infiltrated into the ground with a seepage rate of 3 l/s. The major amount of the tracer was detected at the Gallusquelle, except a small proportion found in the Ahlenbergquelle (Veringendorf), approximately 4.5 km south of the Gallusquelle. Fig. 3 shows the resulting tracer breakthrough at the Gallusquelle. The relatively low discharge during the experiment contributed to a flow velocity of 32 m h^{-1} (dominant tracer velocity), which is low compared to other tracer tests in the catchment (Merkel, 1991). The low total tracer recovery may also be caused by this factor.

3.2 Spring Response to Overflow

During a summer period of five months, five overflow events were observed. Each heavier rainfall event with distinct changes in the spring chemistry and biology was also associated to an overflow of the rain spillway basin. Hence, no data of a spring response to storm events without overflow is available. This paper focuses on event #2, which was analysed in most detail. Heinz (2006) additionally provides information about the other events. The overflow event #2 resulted from a technical failure at the rain spillway basin. Thus, it occurred after a relatively weak recharge event, which caused only a slight increase in discharge at the karst spring. As a consequence, physicochemical changes observed at the spring can be expected to be mainly caused by the sewage infiltration near the rain spillway basin. It should be noted that the precipitation data of one meteorological station cannot represent the recharge conditions of the whole catchment. Additionally, the daily amount of precipitation does not show the intensity of the storm events, which has a strong influence the overflow behaviour of the rain spillway basin. However the data can give an indication, where the strongest responses have to be expected.

3.2.1 Changes in Discharge and Physicochemical Parameters

The rain event causing the overflow on July 25 (Fig. 4) was the first significant event after a two-month period of low groundwater recharge. Spring discharge shows an increase which is only a slight deflection from the continuously decreasing limb that began after event #1, suggesting a low recharge proportion for this event. As a consequence, water temperature did not change as observed after the other events, where temperature decreased significantly by 0.1 to 0.2 K. The rise in discharge began immediately after the overflow, and 24 h later the maximum was reached. Electrical conductivity initially showed a significant increase. Following Williams (1983) this suggests that there is a large proportion of subcutaneous water preceding the event water, which is represented by the relatively low depletion of electrical conductivity. Turbidity showed two small peaks approximately 12 and 48 h after the overflow, each followed by a complete decrease to background level. Both signals arrived before theelectrical conductivity started to increase, i.e. before the subcutaneous water reached the spring. Following Sauter (1992) these peaks are supposed to represent particulate matter from the aquifer itself, resuspended due to higher energy of the transport water. The major turbidity peak is observed along with the depletion of electrical conductivity, suggesting that it consists of particulate matter brought into the aquifer with the infiltrating event water. The following rain events (July 29 and August 2) were not strong enough to cause an overflow, and recharge over the whole catchment was evidently not enough to cause a spring response.

For overflow event #2, the other overflow events, and the artificial tracer test, the dominant flow velocity of the infiltrated wastewater was calculated based on the time interval between the overflow / injection and the maximum contamination / tracer concentration. It was found that velocity and discharge correlated well, following a linear relationship. This suggests that the cross-sections of flow are approximately equal in either case, thus supporting the assumption of an identical contamination source, i.e. the rain spillway basin.

3.2.2 Microbiological Parameters

Although event #2 shows relatively small changes in discharge and physicochemical parameters, the response of the microbiological parameters at the karst spring is significant and its magnitude similar to that of the other events. A likely explanation for this observation is that discharge and physicochemical parameters represent the areal infiltration of event water, i.e. recharge, which is relatively low for event #2, whereas the microbiological parameters are a better indicator of sewage infiltration

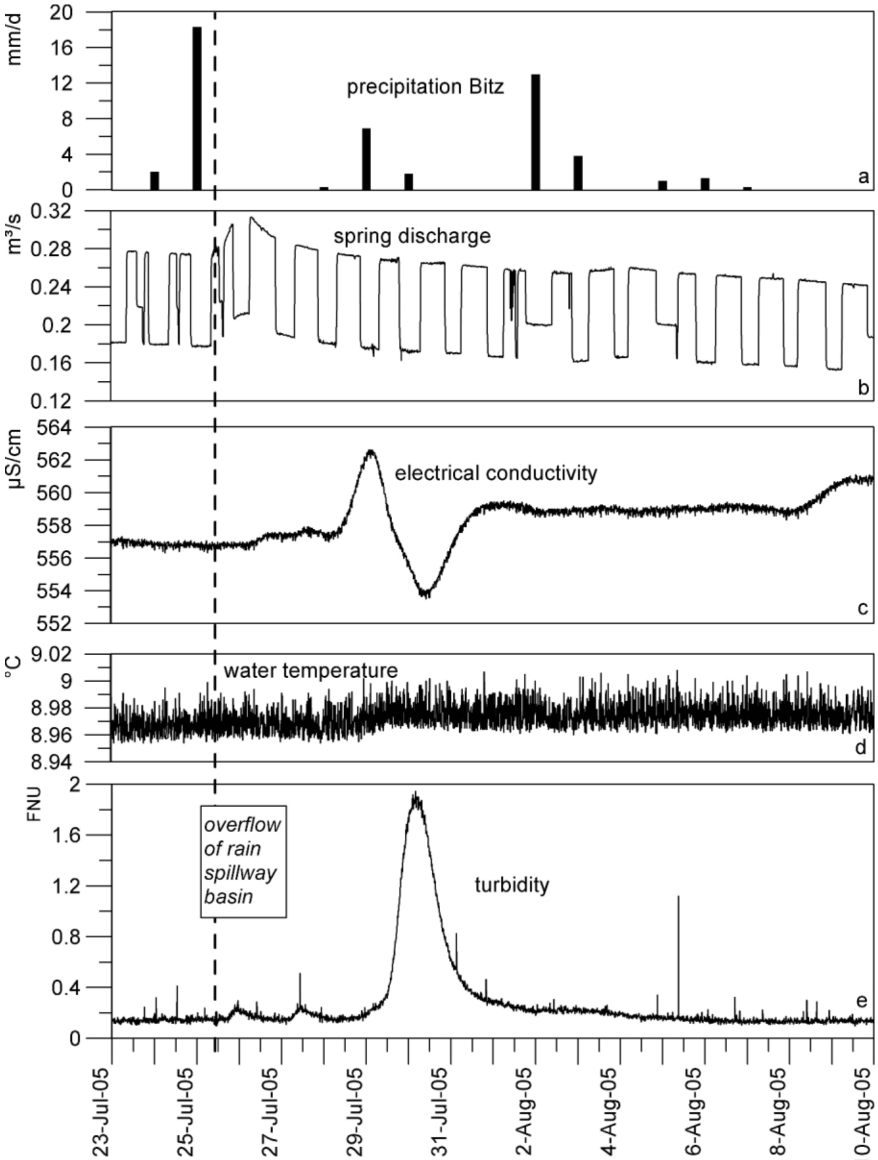

Figure 4: Rainfall, discharge, and physicochemical parameters from 23-Jul to 9-Aug-05 (overflow event #2); (a) precipitation in Bitz, (b) spring discharge, (c) electrical conductivity, (d) water temperature, (e) turbidity at Gallusquelle spring. Dashed line indicates time of overflow. Daily fluctuations in spring discharge are due to water abstraction by the waterworks at night.

from the rain spillway basin. The highest values of microbiological parameters were measured along with the highest turbidity values and the depletion of electrical conductivity (Fig. 5). Subsequently, the recovery of bacterial concentrations to background values developed in a different way than e.g. electrical conductivity: Within 12 h after the peak the number of colonies depleted by 60-70% and *E. coli* by 75%, but over the next ten days the decrease was slow. Turbidity also showed a rapid depletion forming a symmetrical peak in the upper part and then a decrease sustained over the next six days (until August 5). The curve for the total number of colony-forming bacteria resembles the turbidity curve until August 3. Afterwards some single elevated numbers of colonies were measured. Taking into account the standard deviation for the counted numbers on each plate the increase is significant and might be caused by the rain vents that occurred after the overflow (Fig. 4a). The source could be the input of new environmentally adapted bacteria, e.g. via sinkholes, or remobilised germs from the aquifer. Turbidity did not oscillate like the number of colonies suggesting a fraction of free bacteria that do not significantly contribute to turbidity.

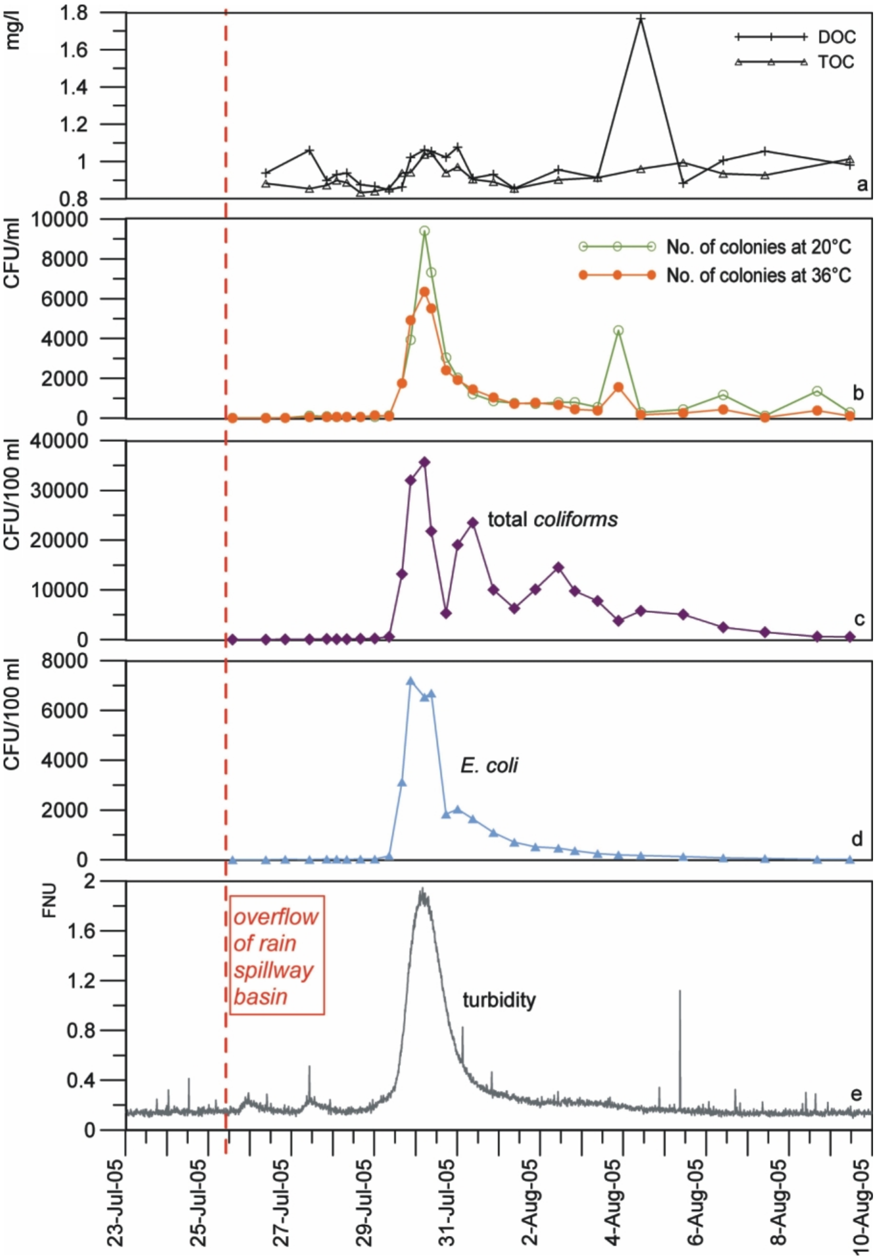

Figure 5: Organic carbon and microbiological parameters from 23-Jul to 9-Aug-05 (overflow event #2); (a) concentration of total and dissolved organic carbon, (b) no. of colonies at 20 and 36°C, (c) total coliformic bacteria, (d) E. coli, (e) turbidity at Gallusquelle. Dashed line indicates time point of overflow; CFU = colony forming unit.

At maximum the *E. coli* concentration reached almost 10,000 CFU/ 100 ml (Fig. 4d), with the highest value measured 8 h before the peak in the total number of colonies, but the concentration stayed at a high level during the breakthrough of turbidity and total number of coloniesat 20 °C and 36 °C, so there is no significant difference in transport behaviour. The continuous and slow decrease following the sharp initial drop can be considered as the steady retardation by the aquifer. In the case of enteric bacteria in the environment this means die off. A particular concentration pattern is shown by the curve of the total coliform bacteria (Fig. 5c). Until the major peak it resembles the curve of the number of colonies at 36 °C and of *E. coli*, whereas the falling limb is interrupted by three intermediate maxima, decreasing one by one, with all concentrations above 2,000 CFU/100 ml. Methodological irregularities can be excluded as a reason (no correlation with sampling time / time lag until laboratory analysis / sample volume) and the changes significantly exceed the standard deviations of the concentrations of total coliforms. Therefore different explanations with regard to ecological or hydrogeological factors were attempted: (a) Coliform bacteria contain one or more groups of bacteria able to proliferate after the input to the aquifer, at least during the first period. This happens batch-wise, so that after the major breakthrough "new" bacteria get discharged. The conditions in sewage contaminated groundwater might benefit their growth. (b) Different transport pathways in the conduit, i.e. the fast flow system, exist that are used by the coliforms. The bulk of bacteria is discharged via the most direct pathway whereas smaller portions arrive later at the spring because of the longer distance of their transport pathway.

Besides a spatial partitioning mechanism there could also be a qualitative separation of bacterial groups due to their size or cell surface properties (charge, affinity to aggregate), resulting in adsorption on the solid phase of the aquifer system. Since longer residence times benefit retardation cell numbers are depleted with time. (c) The latter process plays a role for the following: Water of new recharge events remobilises fractions of coliform bacteria that got adsorbed somewhere on the transport pathway. The high load of nutrients made them able to survive for a certain period. They get discharged until the aquifer is more or less "cleaned". No corresponding signals in spring discharge support this assumption. For both (b) and (c) is valid: As there is little known about special size or affinity to aggregation within the coliform bacteria this does not explain why the effect is limited to this group of bacteria. However, the irregularities in the spring response could be an effect of a combination of the described processes.

Under hygienic aspects, event #2 played an important role because of a disproportionately high input of sewage. The limit of 100 CFU/ml in terms of drinking water requirements for the total number of bacteria at 20°C and 36° C was exceeded by two orders of magnitude. At the end of the measuring period (15 d after overflow) the concentration oscillated around the limit value. High levels of pollution by *E. coli* and total coliforms, even in terms of raw water, were determined during more than 10 days, including the peak concentration of *E. coli* of almost 8,000 CFU/100 ml. At the end of the measuring period the concentration of *E. coli* and total coliforms still indicated too high levels of faecal contamination.

3.2.3 Organic Matter, Major Ions, and Organophosphates

The measurements of the content of total (TOC) and dissolved organic carbon (DOC) apparently had some problems, because DOC yielded sometimes higher values than TOC. Since both values were analysed from two different water samples (filtered for DOC, unfiltered for TOC), the sampling mode could be one source of error. Also the storage period (3 days to 10 weeks) before the measurements could influence the results due to microbial degradation of organic material. If the DOC values of July 27 and August 4 are neglected, a broad peak parallel to the breakthrough of turbidity and bacteria appears (Fig. 5a). This is plausible because bacteria cells as well as a certain fraction of turbidity contribute to the organic content of the water.

Details about major ion concentrations measured during event #2 as well as the other events can be found in Heinz (2006). Due to the dilution effect of the event water calcium, magnesium, and bicarbonate show depletions along with the depletion of electrical conductivity and the major turbidity peak. In contrast, sodium and chloride show a positive deviation from their background level during the turbidity breakthrough. Assuming that these ions originate from the overflow of the rain spillway basin, the total volume of overflow water discharged at the spring can be estimated by mass balance calculations (e.g. Dreiss, 1989).

Applying this mass balance to the measured concentrations of sodium and chloride over the event #2 period yields a maximum proportion of 45% of overflow water in the total spring water along with the contamination peak, which was parallel to the peak in the concentrations of sodium and chloride. Although this number might be an overestimate, it supports the assumption derived from the discharge, turbidity and microbiological data, that the water quality changes were mainly a response to the overflow. Evaluating the sodium concentrations yields a total volume of overflow water of 13,000 m^3, while the chloride concentrations suggest a volume of 73,400 m^3. A comparison with a volume estimate (approximately 23,000 m^3 per overflow) derived from statistical values about the rain spillway basin shows that the estimation provides reasonable results, particularly when the calculation is based on the sodium (Fig. 6).

The difference between the volume estimates resulting from sodium and chloride concentrations is probably due to cation exchange processes reducing the sodium concentrations. However, there might also be additional sources for chloride. In fact, the existence of other sources for sodium cannot be excluded as well. Thus, it was attempted to identify other, more specific anthropogenic contaminants that could be expected to be present in the sewage from the rain spillway basin. To this end, certain synthetic organic compounds, i.e. organophosphates, which are mostly used as flame retardants, plasticisers and lubricants, were quantified around event #5 both in the rain spillway basin and in the spring water. Although the measurements for the Gallusquelle samples yielded very low values, a dependency on the event response was observed, particularly for some compounds, e.g. TCPP (tris-(2-chloro-, 1-methyl-ethyl)-phosphate), TnBP (tri-n-butylphosphate) and TCEP (tris-(2-chloroethyl)-phosphate). The date of the highest values corresponds to the E. coli maximum, indicating the waste water origin of both parameters. Details can be found in Heinz (2006).

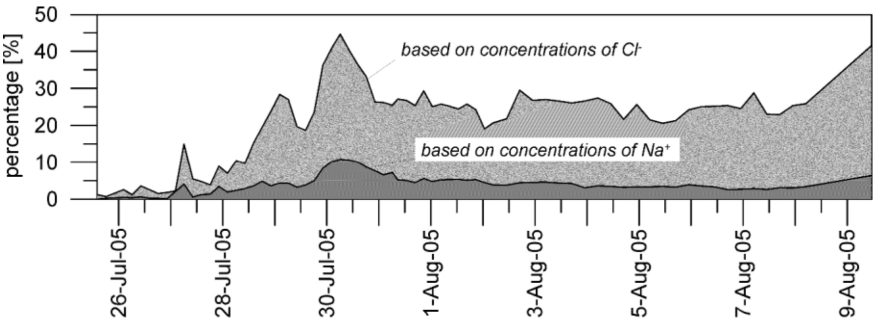

Figure 6: Estimated proportion of overflow water in total spring water for the breakthrough curve of event #2 based on a mass balance of sodium and chloride, respectively, measured in samples of the spring water and once of the filled rain spillway basin.

4. CONCLUSIONS

Five overflow events of different intensity with their consequences at the spring were followed within a period of five months. Each event resulted in a considerable deterioration of the spring water quality, demonstrated by increases in both turbidity and in concentrations of bacteria indicating faecal contamination. This paper has focused on event #2, which resulted from a technical failure after a relatively weak recharge event. Thus, spring response is probably mainly caused by the sewage infiltration near the rain spillway basin. The total number of bacteria showed maxima in the range of 10^3 to 10^4 CFU/ml exceeding the drinking water limit (100 CFU/ml) by one to two orders of magnitude. The number of coliform bacteria showed the strongest deviations from background concentrations (approx. 17 CFU/100 ml), which had been determined during the dry period before overflow event #2. Also the concentration of *E. coli*, which indicates faecal contamination by humans or endothermic animals, rose considerably after overflow events (background approx. 1 CFU/100 ml). The maxima for both parameters were about three to four orders of magnitude higher than the background. Since the German Drinking Water Ordinance requires the absence of coliform bacteria and *E. coli* in a water sample of 100 ml, hygienic standards were missed by far. The results demonstrate the severe effects of spring water contamination caused by the overflow of the rain spillway basin. To avoid such unfavourable conditions, the waterworks are able to stop water abstraction for a short time. However, elevated concentrations of contamination parameters were observed over periods ranging from several days to more than two weeks. Thus a more careful water treatment, e.g. by enhanced ozonation, has to be extended over a longer period resulting in higher treatment costs.

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