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Spatiotemporal scales of river-groundwater interaction – The role of local interaction processes and regional groundwater regimes

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Abstract

Drinking water production in the vicinity of rivers not only requires the consideration of different spatiotemporal scales and settings of river-groundwater interaction processes, but also of local and regional scale groundwater regimes.

Selected case studies in combination with field-experiments and the setup of high-resolution groundwater flow models enabled the investigation of the spatiotemporal development of microbial (classical fecal indicator bacteria and total cell counts) and selected organic micropollutants in riverine and regional groundwater for different hydrological settings, including low and high flow conditions. Proxy indicators suitable as surrogates for the diverse contaminations in alluvial aquifers with different settings could be identified.

Based on the study results, the basic elements for both groundwater management and river restoration concepts are derived, which include the: (1) compilation and evaluation of the "current state" concerning hydrogeology, microbiology and contamination by organic

micropollutants, (2) definition of field-experiments to qualitatively assess variability related to the "current state", and (3) quantitative assessment of groundwater regimes, including variability of groundwater components and inflow areas, by application of high-resolution groundwater flow models.

The validity and transferability of the concept and inferred controls (specifically drivers and controls of river-groundwater interaction) are tested by evaluations derived from hydraulic relationships to river sections with comparable settings and regional groundwater flow regimes in general.

The results of our investigations illustrate the influence of dynamic hydrologic boundary conditions on river-groundwater interaction and of regional scale groundwater flow regimes on the water composition of riverine groundwater systems. It is demonstrated how to identify river sections and their variations with intensified river-groundwater exchange processes and how to quantify the transient character of the different groundwater components that constitute the raw water quality of drinking water wells near rivers.

Keywords: spatiotemporal scales and settings; microbiology; organic micropollutants; filter-efficiency; variability of groundwater components and inflow areas; river sections

1. Introduction

Alluvial aquifers supply a substantial amount of drinking water in many countries. In contrast to common assumptions of many groundwater protection concepts, groundwater recharge often comprises large fractions of river water infiltration (Huggenberger and Epting, 2011). However, regional groundwater flow regimes are also an important component of overall water budgets (Huggenberger et al., 2013). Although groundwater surface water exchange fluxes have been subject to intensive investigations in the last two decades (e.g. Bencala (1993); Cardenas and Wilson (2006); Cardenas et al. (2004); Conant (2004); Kasahara and Wondzell (2003); Wondzell (2006)), the identification of spatial patterns and temporal dynamics of exchange fluxes between groundwater and surface water remains a challenge (Krause et al., 2011; White, 1993).

Exchange fluxes over the river-groundwater interface are controlled by (1) the hydraulic head gradients between groundwater and surface water as a driving force and (2) the hydraulic conductivity of the streambed and aquifer sediments, which control and limit the exchange. The character of these river-groundwater interaction processes is transient and mainly controlled by the discharge dynamics of the surface water. Thereby, the level of the river-head is the decisive factor determining the spatiotemporal constitution of river-groundwater interaction processes.

At the stream-reach to sub-catchment scale, exchange fluxes between groundwater and surface water can be strongly affected by the geological settings, which can be described by the "river corridor" concept introduced by Stanford and Ward (1993). Successively, this concept was further developed in order to consider heterogeneities in the alluvial aquifer and the resulting groundwater flow field (Cardenas and Wilson, 2007; Engdahl et al., 2010; Fleckenstein et al., 2006; Huggenberger et al., 2013).

At smaller, stream-reach scales, however, exchange fluxes over the aquifer river interface appear to be also strongly controlled by spatial patterns of streambed hydraulic conductivity (Calver, 2001; Genereux et al., 2008; Kaser et al., 2009; Leek et al., 2009; Rosenberry, 2008) and streambed topography (Boano et al., 2006; Boano et al., 2010; Cardenas, 2009; Cardenas et al., 2008; Huber et al., 2013; Kasahara and Hill, 2008; Storey et al., 2003; Tonina and Buffington, 2007).

For all scales, quantitative approaches are required to understand how hydraulic fluctuations influence groundwater flow regimes. However, only in a few studies regional flow was considered as an important component of riverine groundwater management (Bencala, 1993; Fleckenstein et al., 2008; Huggenberger et al., 2013; Lewandowski et al., 2009; Schirmer et al., 2014; Ward, 2016). Field-experiments as well as monitoring and modelling approaches were limited to areas located between the drinking water well and the river. Furthermore, numeric analyses mainly were steady state, whereas homogeneous and isotropic conditions were assumed.

Concerning contaminations of aquatic systems by organic MP, most existing sampling strategies focus mainly on the determination of contaminations entering continuously through municipal wastewater treatment plants. Though there have been several studies that describe organic MP for selected rivers and groundwater systems (Benotti et al., 2012; Huntscha et al., 2012; Storck et al., 2012) there are few field studies which relate contaminations to situations during high discharge events (Hillebrand et al., 2012; Hillebrand et al., 2015; Huntscha et al., 2013). For the diverse contaminations, proxy indicators were identified which are best used as surrogates in alluvial aquifers with different settings.

As the detection of many of the waterborne pathogens requires complex analytical procedures (Brookes et al., 2005), *Escherichia coli* (*E. coli*) and Enterococci are generally used as fecal indicator bacteria for microbial pathogens in drinking water (Pronk et al., 2007). However, the vast majority of aquatic bacteria are not pathogens or fecal indicator bacteria, and measuring microbial dynamics in aquatic systems such as groundwater at high frequency is challenging due to the limitations of the conventional, cultivation-based microbial methods. As an alternative, fluorescent staining coupled with FCM is often used for the monitoring, quantification and characterization of bacteria in aquatic ecosystems including freshwater (Gregori et al., 2001), groundwater (Besmer et al. (2016), drinking water (Vital et al., 2012) and wastewater (Foladori et al., 2010). In this study, conventional FCM was applied for monitoring of temporal microbial dynamics in the groundwater systems and the extraction wells.

Because of the often high hydraulic conductivities of the gravel deposits and associated high productivities, drinking water wells often are located close to rivers. As a consequence of this vicinity, such wells are prone to contamination particularly during river flood events. Therefore, two main questions with regard to river-groundwater interaction processes and riverine drinking water protection issues for selected drinking water wells in the Canton Basel-Landscape (Switzerland) were asked: (A) How to assess the “current state” of groundwater bodies related to the variability of groundwater flow regimes and river-groundwater interaction processes as well as microbial and organic MP contaminations, including filter-efficiency and dilution effects? (B) How to localize and quantify the transient character of river groundwater exchange processes for individual river sections as well as the variability of inflow areas and groundwater components of drinking water wells near rivers?

In order to gain a deeper understanding of river-groundwater interaction processes in the context of regional groundwater flow regimes, this study presents a combination of quantitative approaches together with an evaluation of contaminants including microbiological parameters and selected organic micropollutants (MP). The entry points and paths of contaminants into and through the aquifer as well as temporal patterns of the interacting aquifers and surface waters are of utmost importance in increasing the chance of detecting contaminations *insitu*. Several river-groundwater interaction settings are discussed by means of (1) groundwater and river water hydraulics as well as operational regimes of drinking water supplies; (2) analysis of organic MPs, including pharmaceuticals, pesticides, biocides, drugs, sewage tracers, personal care products and industrial chemicals, which are representative of river-groundwater interaction processes and groundwater quality issues; and (3) selected fecal indicator bacteria, and Total Cell Counts (TCC) derived from flow cytometry (FCM) measurements as indicators for hygienic deterioration of water quality.

2. Settings of Case-Study Areas

The study areas are located in the lower Birs, Ergolz and Frenke valleys in Northwest Switzerland (Fig. 1). The investigated settings include: connected river and groundwater systems, including river sections with predominantly infiltration or exfiltration, disconnection of rivers and groundwater systems by a variable thick unsaturated zone as well as transitional systems (Tab. 1). All three locations are characterized by alluvial aquifer systems. The aquifer material of the Quaternary valley fills consists of sandy, variable sorted carbonate gravel (Triassic and Jurassic rock components) with intercalations of silt and clay layers of variable extent, resulting in a large variance of hydraulic properties. All three rivers were canalized at the end of the 19th century and the riverbed was disconnected and lowered several meters below the former floodplain. Further information on the three field sites is given below.

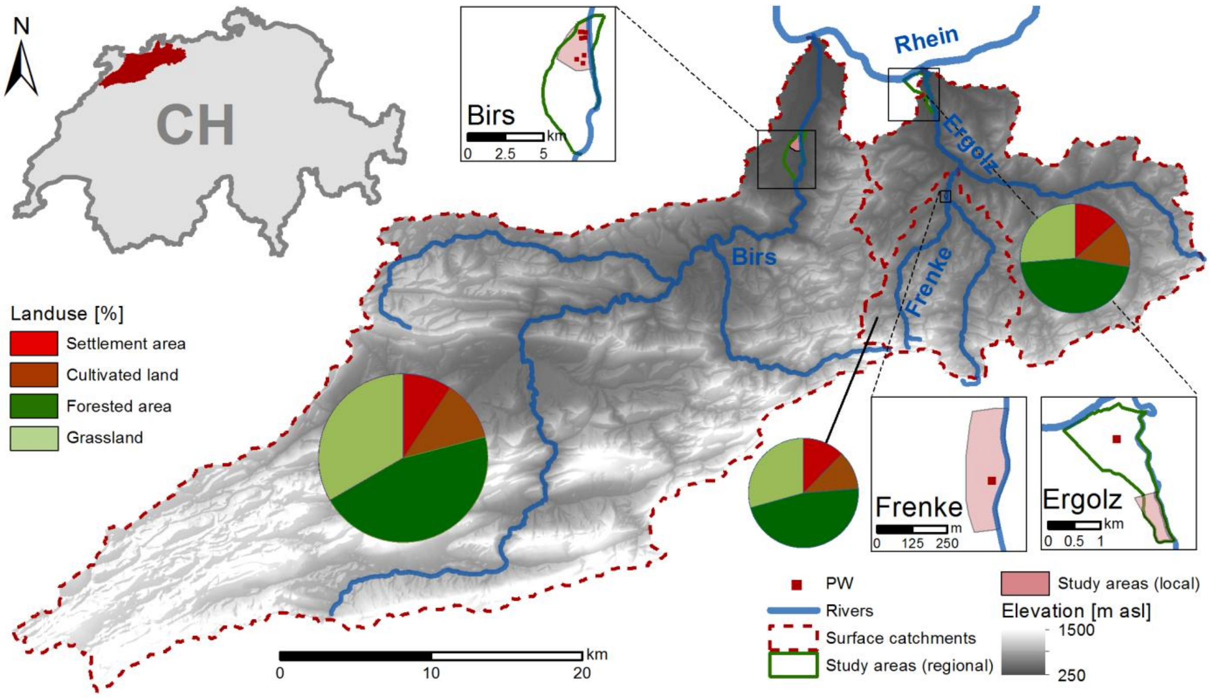

Fig. 1 Location of the three study areas, including the catchment areas of the rivers Birs, Ergolz and Frenke and land use in the different catchments (Data basis: DHM25; SwissTopo (BAFU; BAFU; BFS; BFS)).

Table 1 Spatial and temporal settings for river groundwater interaction processes.

Interaction process	Spatiotemporal mechanism (Hydrogeological & technical settings)
River water infiltration Gradient always from the river to the groundwater table *(connected, disconnected & transitional)*	*Connected:* Continuous infiltration of river water directly into the groundwater saturated zone *Disconnected:* Continuous infiltration of river water into the unsaturated zone and percolation into the groundwater saturated zone *Transitional:* Continuous infiltration for connected and disconnected state in dependence of groundwater table *Superimposed / amplified by groundwater extraction near to the river and / or regional groundwater flow regime (karst and fractured rock aquifer)*
Groundwater exfiltration Gradient always from the groundwater table to the river *(connected)*	*Connected:* Continuous exfiltration of groundwater into the river
River water infiltration Gradient from the river to the groundwater table only during flood events *(connected)*	*Connected:* Infiltration of river water directly into the groundwater saturated zone only during flood events when the river stage temporary rises above the groundwater table
Groundwater exfiltration Gradient from groundwater table to the river only during flood events *(connected & disconnected & transitional)*	*Connected:* Exfiltration of groundwater directly into the river only during flood events when the gradient of the regional groundwater temporary rises above the river stage (pressure propagation)
River water infiltration Gradient from the river to the groundwater table only between flood events (connected & disconnected & transitional)	*Connected / disconnected / transitional:* Infiltration of river water directly into the groundwater saturated zone or into the unsaturated zone only between flood events. During flood events the groundwater table temporary rises above the river stage (rapid pressure propagation around bedrock steps or within karst and fractured rock aquifers)

Furthermore, we extend the GroundWater Body (GWB) approach that originally was introduced by the Water Framework Directive Commission (CEC 2000). Our approach considers a GWB as a distinct volume of groundwater that can be effectively managed and for which boundary conditions (hydraulic, thermal, chemical, etc.) can be delineated. Most aquifer systems comprise several isolated or connected GWB.

Birs GWB

The river Birs runs 75 km through the Swiss Jura and joins the river Rhine in the city of Basel, creating a catchment area of 979 km^2 (Fig. 1). The mean annual flow near the confluence zone of the tributary Birs with the Rhine is 15.4 m^3s^{-1} (flow depth approx. 1.27 m) and storm flows can reach up to 383 m^3s^{-1} (flow depth approx. 4.35 m; Swiss Federal Office for the ENvironment FOEN-station 2106).

At the study location the aquifer bottom is formed by an aquitard consisting of Tertiary deposits of Elsässer Molasse (sandstone marl bedrock) with very low hydraulic conductivities. The thickness of the saturated zone varies between 0.6 and 10 m. The recharge of the GWB is controlled by regional groundwater flow components and river water infiltration, lateral inflow from the local catchment area and intermittent artificial recharge (Affolter et al., 2010b).

Eight active Pumping Wells (PW) in the study area (Fig. 2) supply 5E06 m^3 drinking water per year to six communities, comprising approximately 51,000 people. Land use in the catchment area comprises approx. 9% urbanized surfaces, including traffic lines and repositories, 12% cultivated land, 46% forested areas and 33% grassland (BAFU) (Fig. 1). Estimations of the fraction of sewage water (derived from population equivalents) in relation to annual river discharge total to approx. 3.5% (BAFU).

Ergolz GWB

The river Ergolz runs 28 km through the Swiss Jura and joins the river Rhine in the municipality of Augst, creating a catchment area of 299 km^2 (Fig. 1). The mean annual flow of the Ergolz measured approx. 6 km upstream of the confluence zone with the Rhine is 3.7 m^3s^{-1} (flow depth approx. 0.51 m) and storm flows can reach up to 155 m^3s^{-1} (flow depth approx. 3.27 m; FOEN-station 2202).

At the study location the aquifer bottom is formed by an aquitard consisting of Tertiary deposits of Meletta layers (clay-/sandstone bedrock) with very low hydraulic conductivities. The thickness of the saturated zone varies between 0 and 12 m. The recharge of the GWB is controlled by regional groundwater flow components from the main valley as well as from river water infiltration and lateral inflow from the local catchment area.

Land use in the catchment area comprises approx. 14% urbanized surfaces, including traffic lines and repositories, 14% cultivated land, 46% forested areas and 26% grassland (BAFU) (Fig. 1). Estimations of the fraction of sewage water (derived from population equivalents) in relation to annual river discharge total to approx. 8.5% (BAFU).

Frenke GWB

The river Frenke originates from the confluence of the tributary rivers Vordere and Hintere Frenke in the municipality of Bubendorf. After flowing 18.7 km through the Swiss Jura the river Frenke joins the river Ergolz in the municipality of Liestal, creating a catchment area of 86 km^2 (Fig. 1). The mean annual flow of the Frenke measured near the confluence zone is 1.4 m^3s^{-1} (flow depth approx. 0.36 m) and storm flows can reach up to 52.2 m^3s^{-1} (flow depth approx. 1.64 m; stations 309 and 319 run by the civil engineering department of the Canton Basel-Landscape).

At the study location the aquifer bottom is formed by an aquitard consisting of Jurassic deposits of Hauptrogenstein (carbonates) and Unterer Dogger (layering of marls, claystone and carbonates), with very low hydraulic conductivities. The maximal depth to groundwater is 8.2 m and the thickness of the saturated zone varies between 8.8 and 14.2 m. The recharge of the GWB is controlled by regional groundwater flow components from the main valley as well as from river water infiltration and lateral inflow from the local catchment area. In addition, large scale analysis of tectonic structures (Horst and Graben structures of the Tabular Jura) and the results of hydrogeophysical investigations (Electrical Resistivity Tomography) indicate the existence of several fault systems crossing the study area. Therefore, beside groundwater recharge by infiltrating river water, both, the interaction with the regional groundwater flow and the regional Karst, respectively fault system, have to be considered.

Land use in the catchment area comprises approx. 12% urbanized surfaces, including traffic lines and repositories, 12% cultivated land, 47% forested areas and 29% grassland (BAFU) (Fig. 1). Estimations of the fraction of sewage water (derived from population equivalents) in relation to annual river discharge total to approx. 6% (BAFU).

3. Approach and Methods

Field experiments and monitoring systems

The setup of the various field-experiments and monitoring systems was focused on capturing the spatiotemporal dynamics of river-groundwater interaction processes along the different river sections and within the inflow area of the PW. This includes a selection of river sections according to the requirements for quantifying the interaction processes and capturing relevant qualitative parameters. A special focus was also placed on quantitatively and qualitatively capturing the dynamics of the regional groundwater flow regimes.

A baseline sampling performed in December 2013 for all three study sites during low river discharge and low groundwater table conditions provided an inventory of the current state of water quality on microbial and organic MP contaminations. Furthermore, the baseline sampling served the design of specific field-experiments and the selection of indicator substances. Table A.1 in the appendix summarizes the measurements and experiments performed within the three study areas, including the different monitoring systems, located within the rivers, groundwater Observation Wells (OW) and PW, respectively. Experiments were performed during dry weather conditions and flood events.

Beside conventional OW for the study area located at the river Birs also three OW clusters, each covering three depths of the aquifer was available. For the investigation sites located at the river Ergolz and Frenke at three and two locations, respectively, near the rivers Continuous Multichannel Tubing CMT-systems were installed with the Direct Push (DP) method (Butler et al., 2002; Dietrich and Leven, 2006; Leven et al., 2010) which allows depth-oriented groundwater sampling in three different depths of the aquifer.

Groundwater components, delineation of river sections and inflow areas

An understanding of how water quality is constituted at the sampling locations is only possible when the non-stationary character of the quantity and quality of the different groundwater components can be determined. This knowledge also allows investigating the inflow areas of drinking water wells near rivers and delineating those river sections which are most promising for restoration measures without any major accompanying measures.

By means of calibrated and validated high-resolution groundwater flow models the spatial and temporal settings for river-groundwater interaction processes were investigated (Fig. 2). The models also were used to investigate the temporal composition of groundwater components for any desired location of the modeled GWB. A method is presented which allows deriving groundwater components of different provenance for all three study sites, including:

(A) Groundwater related to the regional groundwater flow system;

(B) River water infiltrated into the groundwater system with short residence times;

(C) Groundwater mostly related to recharge from adjacent hillslopes.

For the quantitative evaluations the three rivers were subdivided into sections according to the information available on river bed topography, whereas individual sections typically are separated by artificial steps in the river bed (Fig. 2).

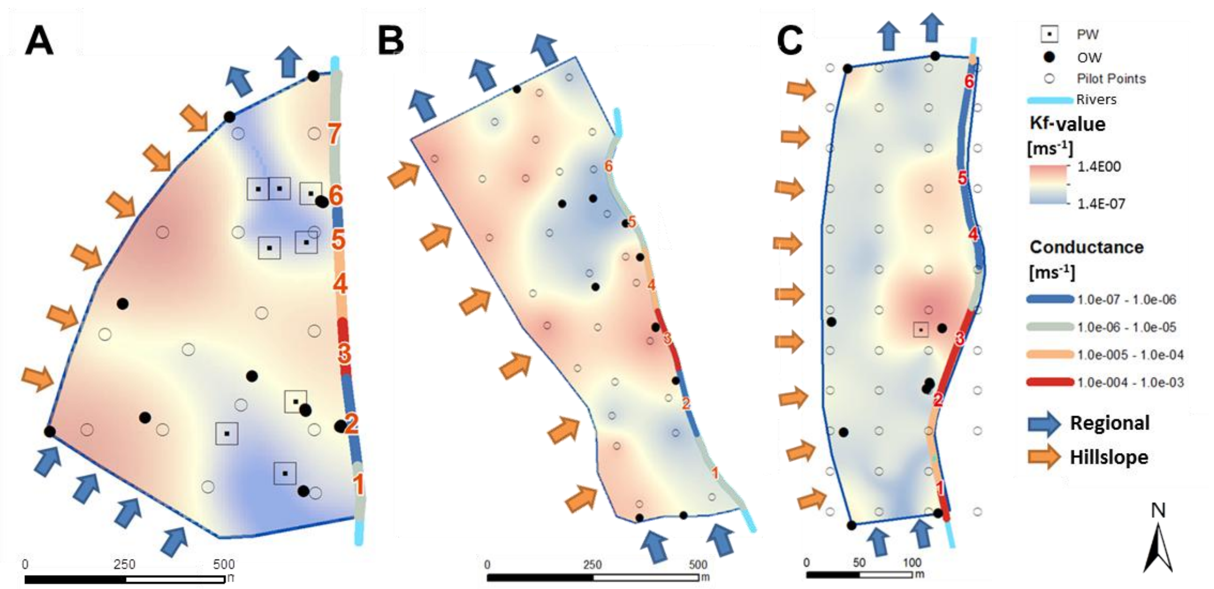

Fig. 2 Calibration results of hydraulic conductivities and river bed conductance of the various river sections for the GWB along the Rivers Birs (A), Ergolz (B) and Frenke (C). Whereas, for the Birs and Ergolz GWB pilot points were introduced manually, for the Frenke GWB an approach with a regular grid of pilot points, including several pilot points outside the modeling domain, was chosen.

Furthermore, the non-stationary character of inflow areas to individual PW and those river sections which are located within the inflow areas were investigated using different hydrogeological and operational boundary conditions. The variability of inflow areas (filter paths) and infiltrating river sections then were ratified by: (1) quantitatively and qualitatively comparing the inflow areas for low and high groundwater and river water levels; and (2) characterizing the infiltrating river section in the inflow area in relation to the river bed topography.

Table A.2 in the appendix summarizes the data sets which were used to set up the model geometries, to parameterize the boundary conditions as well as to calibrate and validate the groundwater flow models.

Microbiology and filter-efficiency

FCM was used to determine the TCC. Measurements and analysis were performed as described in (Prest et al., 2013). In short, 500 µL of each water sample was stained with 5 µL SYBR® Green I (SG, Invitrogen AG, Basel, Switzerland; 100x diluted in TRIS buffer, pH 8). Prior to measurements, samples were incubated for at least 10 min at 37 °C. A BD Accuri C6® flow cytometer (BD Accuri Cytometers, Belgium) was used, applying the same settings and gating strategy as described in (Prest et al., 2013).

For fecal indicator bacteria plating, the standard methods ISO 16649-1 and ISO 7899-2 for membrane filtration and plating methods for the enumeration of *E. coli* and Enterococcus, respectively, were used.

In order to evaluate the filter-efficiency between the origin of infiltration at the riverbed to the corresponding OW or PW, flow lines were simulated with the groundwater flow models. These models were also used to gain information on the temporal settings of the individual experiments, i.e. particles were backtracked to the hydraulic origin of river water infiltration for the individual sampling days.

The filter-efficiency λ of the aquifer was calculated according to Eq. 1 (based on Matthess et al. (1988)) and Eq. 2:

$$\lambda = \frac{\ln\left(\frac{C_P}{C_0}\right)}{x_h}, \qquad (1) \qquad\qquad \text{and} \quad x_h = \sqrt{\Delta h^2 + s^2} \qquad (2)$$

where C_0 is the concentration in the river water, C_P the concentration in the OW and x_h is the diagonal flow length between the river and the individual observation point, where Δh is the height difference between the river board and the middle of the filter section of the individual observation points and s is the length of the simulated groundwater flow line from the observation point to the riverbank.

Organic MP and derivation of indicator substances

Organic MP were analyzed with different methods using for polar compounds off-line SPE (Solid Phase Extraction) coupled to liquid chromatography with detection by high resolution mass spectrometry after electrospray ionization (LC-HRMS, Schymanski et al. (2014)) for screening of hundreds of organic MP and online-solid phase extraction-LC-tandem mass spectrometry (online-SPE-LC-MSMS, Huntscha et al. (2012)) for sensitive detection of a defined organic MP set. The measurement of a defined set of volatile organic compounds (VOC), representative mostly for industrial sources, were carried out by GC-MS (Gas Chromatography - Mass Spectrometry) and an additional GC-MS Screening allowed the detection of unknown volatile compounds. As described above, a baseline sampling in December 2013 was performed at all three study areas during low river discharge and low groundwater tables followed by several sampling campaigns during high discharge events (Table A.1 in the appendix). From the screening with LC-HRMS in 2013 (Hollender et al., in prep.) 14 indicator organic MP were selected, representative for input from agriculture, urban wastewater and industry (Tab. 2) and analyzed in all further field experiments by online-SPE-LC-MSMS. In dependence of the respective organic MP the limits of quantification (LOQ) ranged between 1 and 50 ngL^{-1}.

Table 2 The selected indicator organic MP with physical-chemical properties (EPA, EPI Suite estimate).

Indicator OMP	Explanation	CAS No.	Substance property			Biodegradability	Origin		
			$logK_{ow}$	pKa	Main ioniza-tion at pH 7*		Industry	Domestic	Agriculture
Acesulfame	food additive, sweetener	55589-62-3	-1.3	3.0	a	persistent	-	+	-
Atenolol	pharmaceutical, antihypertensive	29122-68-7	0.2	9.7	c	degradable	-	+	-
Atenolol acid	pharmaceutical, metabolite	56392-14-4	-2.3	9.7	a + c	degradable	-	+	-
Atrazine	pesticide / herbicide	1912-24-9	2.6	3.2	n	persistent	-	-	+
1-H-Benzotriazole	anticorrosive	95-14-7	1.4	8.6	n	mostly persistent	+	+	-
Caffeine	tracer	58-08-2	-0.1	0.9	n	well degradable	-	+	-
Carbamazepine	pharmaceutical, anticonvulsant	298-46-4	2.5	16	n	persistent	-	+	-
Carbamazepine-10-11-dihydro-10-11-dihydroxy	pharmaceutical, metabolite	58955-93-4	-0.2	12.8	n	persistent	-	+	-
Candesartan	pharmaceutical, antihypertensive	139481-59-7	4.8	3.9	a	persistent	-	+	-
Diclofenac	pharmaceutical, analgesic	15307-86-5	4.0	4.0	a	degradable	-	+	-
Hydrochlorothiazide	pharmaceutical, diuretic	58-93-5	-0.1	9.1	n	persistent	-	+	-
Lamotrigine	pharmaceutical, anticonvulsant	84057-84-1	2.6	5.9	n	persistent	-	+	-
Metolachlor ESA	pesticide, metabolite	171118-09-5	1.7	13.7	a	persistent	-	-	+
Terbuthylazine	Pesticide, herbicide	5915-41-3	3.3	2.0	n	persistent	-	-	+

4. Results

In the following the research results are presented and the “current states” as well as the variability concerning hydrogeology, microbiology and contamination by organic MP of the investigated GWB are outlined.

All datasets from the field experiments and monitoring systems were evaluated in context by the results derived from the high-resolution groundwater flow models. Specifically, the micro-biological parameters and filter-efficiencies as well as organic MP and derived indicator sub-stances are discussed in relation to the dynamics of groundwater components at the PW. Additionally, the variability of inflow areas and infiltrating river sections is discussed.

The integration of the different methodological approaches facilitated comparing the different spatiotemporal settings within the three investigation sites in the context of the regional groundwater flow regimes, including different hydrogeological boundary conditions. Table 3 summarizes the knowledge gained and the limitations associated with the applied methods. The table provides information on the necessary clarifications and an overview of the method selection.

Table 3 Methodological approaches including knowledge gain as well as restrictions and disadvantages.

Method	Knowledge gain	Restrictions / disadvantages
Monitoring systems		
cantonal GW*- and SW**-monitoring; canton & FOEN***	base data sets with long-time time series	location, construction and instrumentation not project-specific
extension GW- and SW-monitoring systems RWBL21	completion hydraulic measurements for description of regional and local GW flow regimes; calibration and validation of GW-models	time-series only for project-duration very high personal and support expenses
CMT****-monitoring systems	depth-oriented sampling	operability of single channels (measurement depths); representativeness of measurement results
Field-experiments / laboratory analysis		
standard water analysis	mineralization; definition hydro chemical „current status“	expenditure sampling / analytics; restricted validity concerning process-investigations
plating / cultivation of indicator-bacteria	risk assessment of microbial contamination of OW and GW, classification of results FCM	Time-span from sampling to analytical results; restricted validity of routine sampling procedures
Flow CytoMetry (FCM)	definition of „current status“ microbiology OW and GW; microbial activity	expenditure sampling / analytics; restricted validity concerning pathogens
ICP-MS element analytics	definition of „current status“ inorganic elements	expenditure sampling / analytics; restricted validity concerning process-investigations
stable isotope analysis of ^{2}H and ^{18}O	water age, origin, mixing	derivation of distinct end members; necessary spatiotemporal resolution of sampling
offline Solid Phase Extraction (SPE) and Liquid Chromatography coupled with High-Resolution Mass Spectrometry (LC-HRMS)	quantitative analysis and screening of unknown polar OMP+	expenditure sampling / analytics
online SPE coupled with LC-MS/MS	quantitative analysis of polar OMP	expenditure sampling / analytics
purge-and-trap Gas Chromatography coupled with Mass Spectrometry (GC-MS) and GC-MS screening	quantitative analysis of volatile organic substances and unknown, analysis of volatile organic substances	expenditure sampling / analytics
GW-Modelling		
flow	characterization of GW flow regimes (GW-heads; -budgets; flow velocities; inflow areas to PW++)	expenditure; necessary time-series; uncertainties modeling results
transport	derivation GW-components	uncertainties modeling results
scenario definition	evaluation of operational and hydrological variable boundary conditions; comparison of different restoration measures	extrapolation boundary conditions
Further methods		
filter efficiency	measure for contaminant reduction	reference location of infiltration and flow path to PW
deconvolution	derivation of flow velocities and duration on basis of T+++ or EC- measurements in SW and GW	reference location of infiltration and flow path to PW differentiation of observed EC-changes in PWs (OW-infiltration or different GW-components)

*GroundWater; *** Surface Water; ***Federal Office for the Environment; +Organic MicroPollutants; ++Pumping Well; +++Temperature; -Electrical Conductivity;

Birs GWB

Figure 3A to C illustrates the hydraulic settings of the Birs GWB by means of measured river and groundwater heads in relation to the local elevations of the river bed for the two OW clusters in the inflow areas of PW1 and 2. At the location of the OW1 cluster in the inflow area of PW1 the riverine groundwater level generally was below the river bed for most of the measurement period. The local groundwater table was only connected to the river during one larger flood event (>100 m^3s^{-1}) in December 2013. Succeeding flood events, with even higher discharges, did not result in a connection between the groundwater table and the river. Therefore, for this location continuous river water infiltration into the unsaturated zone and percolation into the groundwater saturated zone was the dominant process (Tab. 1). At the location of the OW2 cluster in the inflow area of PW2 the riverine groundwater level generally was above the river head. For this location therefore, continuous groundwater exfiltration into the river was the dominant process (Tab. 1).

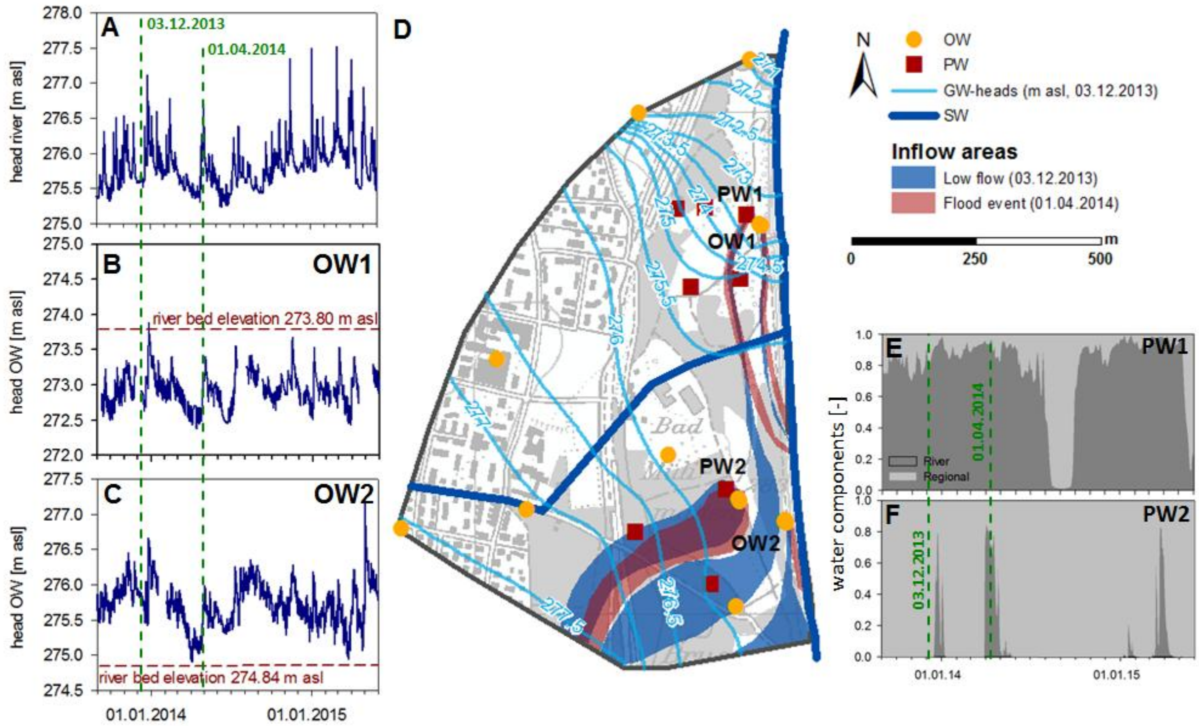

Fig. 3 Groundwater flow regime for the study area located along the River Birs (Fig. 1). A-C: Measured river and groundwater heads in relation to the local elevation of the river bed of the two OW in the inflow areas of PW1 and 2. D: Calculated groundwater heads and inflow areas depicted for a hydraulic situation with low river discharge and low groundwater table (03.12.2013) and inflow areas during a moderate flood event (01.04.2014), dark red: overlay of inflow areas; SW: Surface water. E&F: Calculated groundwater components for PW1 and 2. Dotted vertical green lines mark the studied hydraulic situations.

Figure 3D illustrates the variability of inflow areas (filter paths) and the different infiltrating sections along the river during a hydraulic situation with low river discharge (12.7 m^3s^{-1}) and low groundwater table (03.12.2013) as well as during a moderate flood event in an extremely dry time-period (6.7 m^3s^{-1}; 01.04.2014). During hydraulic settings with low river discharge and low groundwater table the inflow areas to PW1 are oriented towards the regional groundwater flow regime to the south. During flood events the inflow areas to PW1 as well as OW1 cluster progress approx. 300 m parallel to the river before they orient towards it. The inflow area to the OW1 cluster always is oriented towards the river whereas the connected infiltrating river section shifts to the north during elevated river discharges. For the studied hydraulic settings the inflow area to PW2 always is oriented towards the regional groundwater flow regime to the south. During hydraulic settings with low river discharge and low groundwater table the inflow area to the OW2 cluster are oriented towards the regional groundwater flow regime to the south. During flood events the inflow area to the OW2 cluster progress approx. 50 m parallel to the river before it orients towards it. As a result of elevated hydraulic gradients during and after flood events the inflow areas are narrower compared to settings with low hydraulic conditions.

Figure 3E and F show the calculated water components that constitutes the raw water of PW1. During average hydraulic settings the raw water is constituted by approx. 80% by infiltrated river water with short residence times (hours to days); only approx. 20% can be related to regional groundwater components. In August/September 2014 as well as June 2015 situations were observed where the fraction of regional groundwater components can reach 70 to almost 100%. Such situations result when the regional inflow to the investigated GWB is comparably high and river infiltration, related to the southern river sections, is reduced. On the contrary, during average hydrological conditions the raw water of PW2 is constituted by approx. 90% which can be related to regional groundwater components; only approx. 10% can be related to infiltrated river water with short residence times (hours to days). In December 2013, April 2014, as well as March/April 2015 and January 2015 situations were observed where the fraction of infiltrated river water with short residence times reached 87%, whereas no direct correlation with flood events were found. Moreover, such situations result when the regional inflow to the investigated GWB is comparably low and river infiltration related to the southern river sections is comparably high.

Figure 4 summarizes the results of FCM data for different hydrologic conditions (see Table A.1 in the appendix) measured in the river Birs, the PW1 and 2 as well as for depth-oriented samples of the OW clusters in the inflow areas of the PW. From the river to the PW, the TCC is reduced by two orders of magnitude. Concerning the microbial contamination by indicator organisms, *E. coli* could only be detected within the surface-near OW located in the inflow area of PW2.

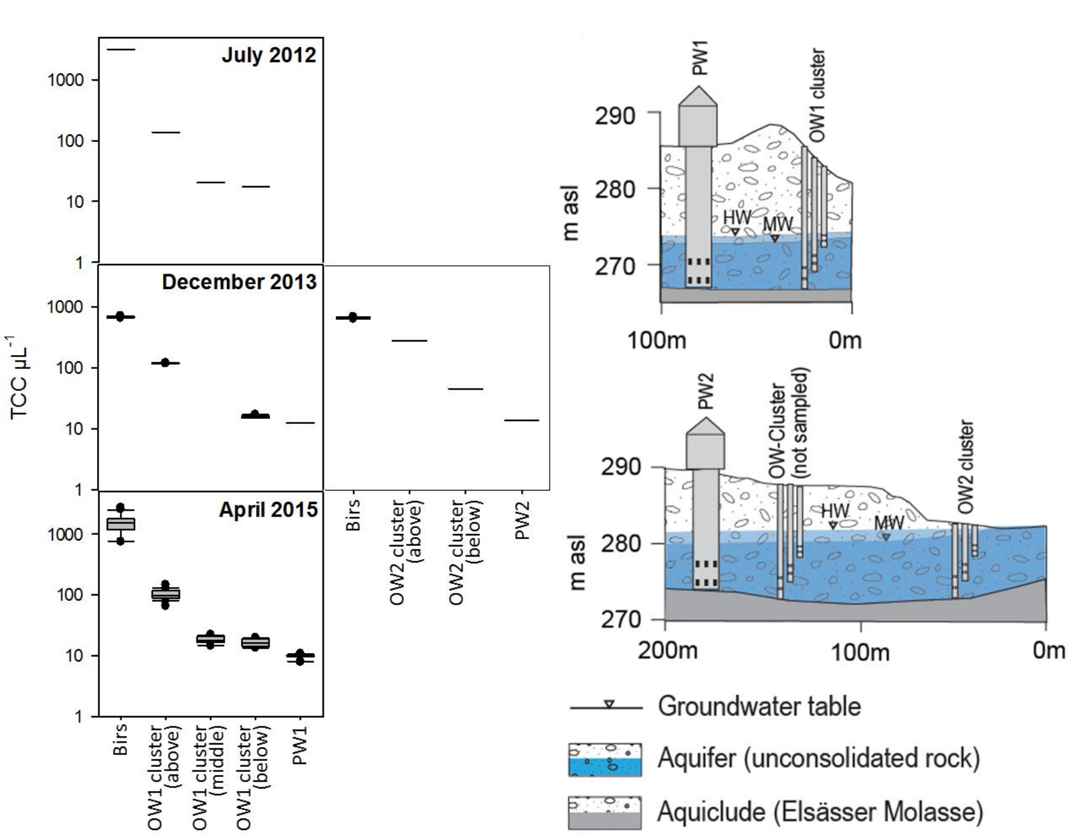

Fig. 4 Left: Illustration of FCM-TCC data for different hydrologic conditions (see Tab. A1) measured in the River Birs, the PW1 and 2 as well as for depth-oriented samples of the OW clusters in the inflow areas of the PW (horizontal bars represent single measurements). Right: Hydrogeological cross section through the aquifer from the river board to the different OW and PW1 (top right) and PW2 (bottom right).

In Figure 5 the results of the indicator organic MP are summarized as boxplots for all measurement campaigns performed at the Birs GWB. The wastewater tracers acesulfame, 1H-benzotriazole and caffeine are found, as expected, in higher concentrations than all other pharmaceuticals and pesticides as well as their transformation products. Concentrations of indicator organic MP differ for groundwater samples taken near the river and for locations which can be related to the regional groundwater flow regime. At the sampling location of the OW2 cluster (top, Fig. 5) near to the river Birs similar concentrations of indicator organic MP as in the river Birs were detected. However, at the sampling location of the OW1 cluster (top, Fig. 4), which is also located close to the river, only for some indicator organic MP increased concentrations were detected. Likewise, acesulfame concentrations are comparable at the sampling location OW1 cluster (top) and in the river water sample. This observation indicates that the entry of organic MP into the GWB not only occurs through the local infiltration of river water but also through the regional groundwater flow regime (e.g., leaky sewers or diffuse sources, see also Fig. 3). It has to be taken into account that the regional groundwater flow is composed to a large extent of river water, which has infiltrated further upstream (Fig. 5). However, as the flow paths and with this, the residence times of this groundwater are much longer in comparison to groundwater from direct bank filtration close to the PW, it is referred to as regional groundwater. The concentrations of all organic indicator MP, apart from acesulfame, are on average less than 250 ngL^{-1}. Variations in concentrations can be attributed to fluctuations in the hydrological conditions and waste water entries. Diclofenac, which behaves relatively persistent during the passage through sewage treatment plants (Tab. 2), is only detected in significant concentrations above 5 ngL^{-1} in the river Birs. In the groundwater samples it was only detectable in traces, since it is rapidly degraded or transformed during river bank filtration (Huntscha et al., 2013; Schymanski et al., 2014). Atenolol acid, the transformation product of atenolol, was detected in the river Birs, two PW and one OW next to the river, indicating younger groundwater with short residence times or a locally poorer degradation potential. The remaining indicator organic MP, which are classified as difficult to decompose and have little sorption capacities due to relatively low log K_{ow} values and partial occurrence as anions at groundwater pH (Tab. 2), can be detected in some groundwater samples, but in low concentrations. In general, due to biodegradation, sorption during riverbank filtration and dilution with other groundwater components, organic MP concentrations in general are higher in the surface water than in the groundwater samples.

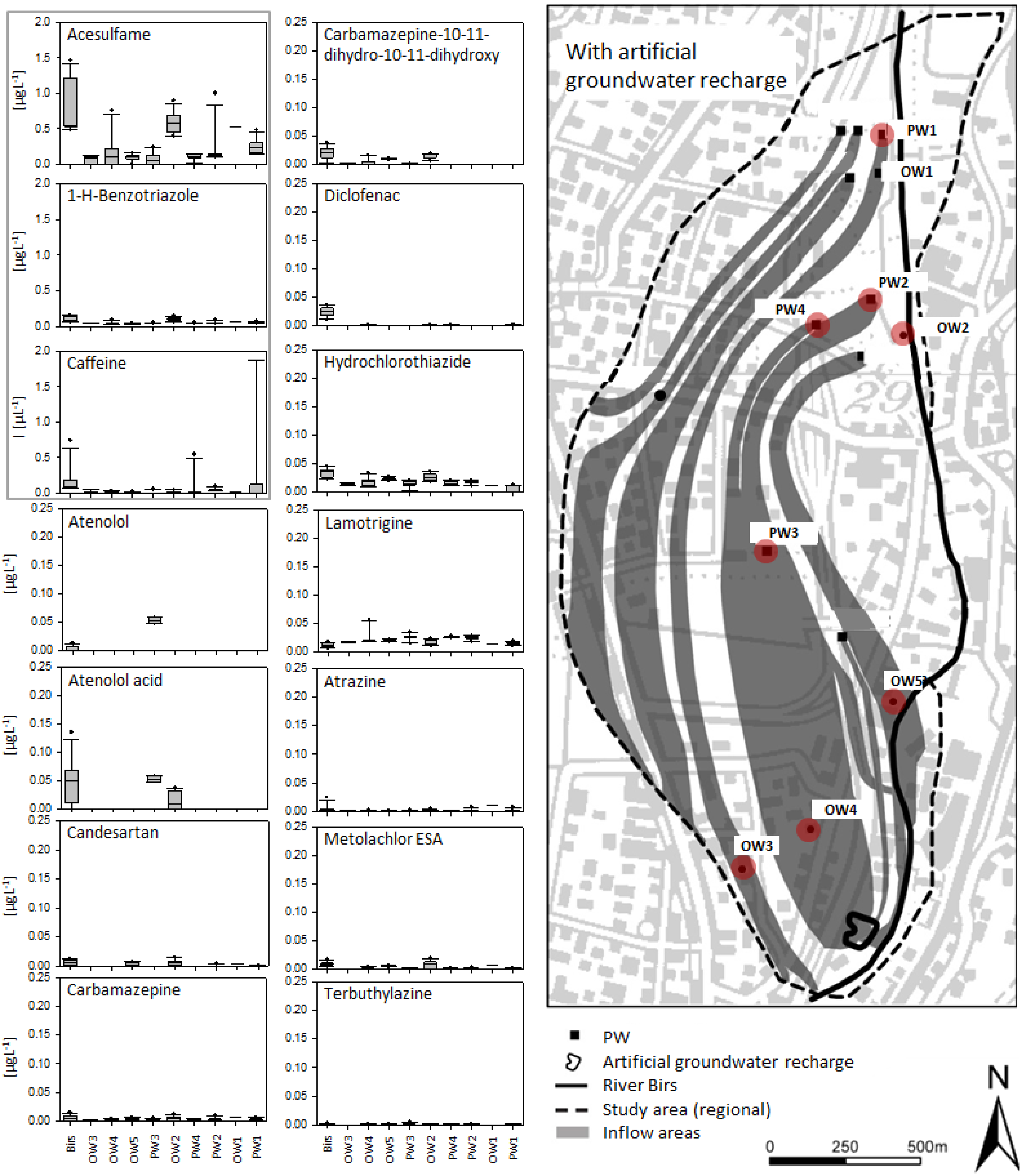

Fig. 5 Left: Descriptive statistics of organic indicator MP (Tab. 2) measured during all campaigns in the Birs GWB (gray box shows tracers with concentration resolution up to 2 µgL^{-1}, all others up to 0.25 µgL^{-1}). Right: Sampling locations (red points) related to the regional groundwater flow regime and within the inflow areas of the PW (modified after (Affolter et al., 2010a).

Ergolz GWB

Figure 6A to E illustrates the hydraulic settings of the Ergolz GWB by means of measured river and groundwater heads in relation to the local elevation of the river bed for four OW close to the river Ergolz. At the location of these OW the riverine groundwater level generally is below the river bed for most of the measurement period. Therefore, for this location a continuous river water infiltration into the unsaturated zone and percolation into the groundwater saturated zone is the dominant process (disconnected interaction type; see Tab. 1).

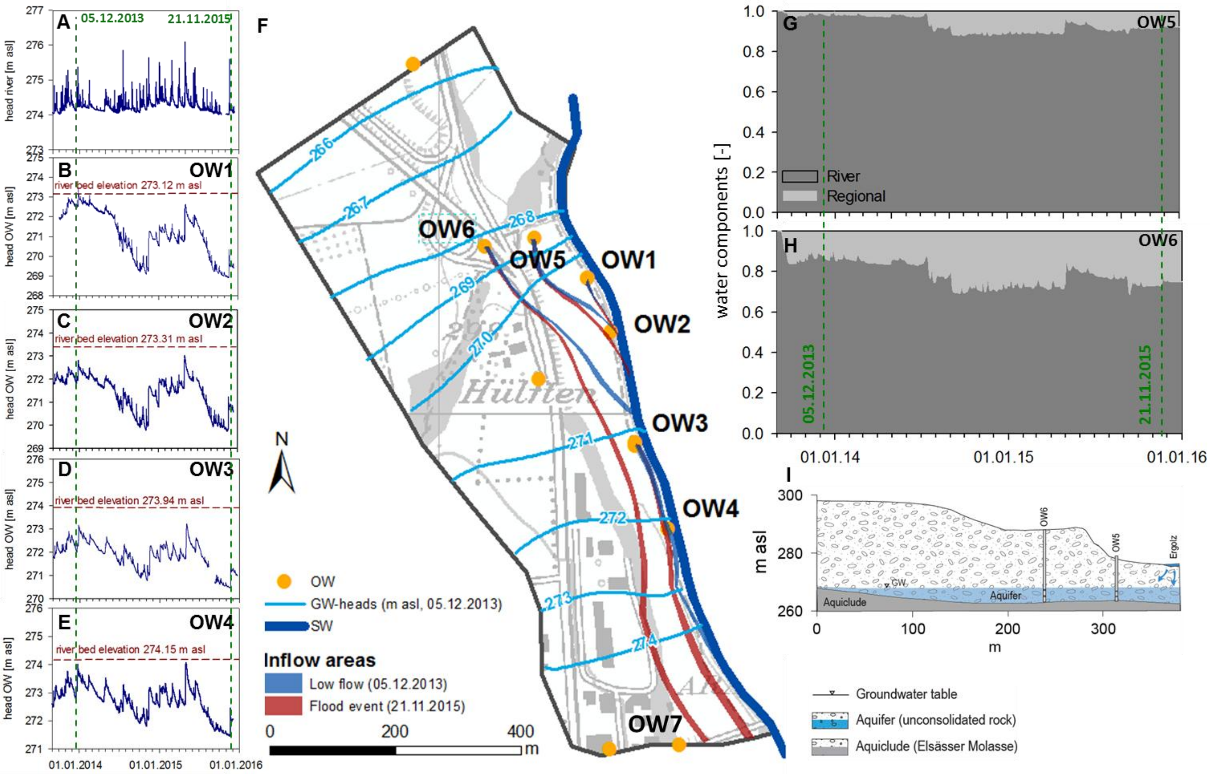

Fig. 6 Groundwater flow regime for the study area located along the River Ergolz (Fig. 1). A-E: Measured river and groundwater heads in relation to the local elevation of the river bed for the four riverine OW. F: Calculated groundwater heads and inflow areas to OW3, 5 and 6 for a hydraulic situation with low river discharge (05.12.2013) and inflow areas during a moderate flood event (21.11.2015); SW: Surface water. G & H: Calculated groundwater components for OW5 and 6. I: Hydrogeological cross section through the aquifer from the river board to the different OW. Dotted vertical green lines in the diagrams A to E, G and H mark the studied hydraulic situations.

Figure 6F illustrates the variability of inflow areas (filter paths) and the different infiltrating sections along the river during a hydraulic situation with low river discharge (2.9 m^3s^{-1}, 05.12.2013) as well as during a moderate flood event (11 m^3s^{-1}; 21.11.2015). During hydraulic settings with low river discharge the inflow areas to OW3, 5 and 6 orient towards the river. In the course of flood events, the inflow areas of OW3 and 6 orient towards the regional groundwater system and the inflow area of OW5 shifts to a river section located approx. 50 m to the south. The inflow areas are narrower when compared to those to the PW located in the investigation area along the river Birs, as no active pumping is performed in the OW located in the investigated area along the river Ergolz.

Figure 6G and H shows the calculated water components that constitutes the raw water at the different OW. During average hydrological conditions the raw water of OW5 and 6 is constituted by approx. 95 and 80%, respectively, of infiltrated river water with short residence times (hours to days); only approx. 5 and 20%, respectively, of groundwater components can be related to the regional groundwater system.

Figure 7 summarizes the FCM data during a moderate flood event in the river Ergolz and the field experiment in November 2015. Concerning the sample in the riverine OW3, it is uncertain whether the maximum microbial contamination was detected as the sampling took place when the hydrograph was already declining. In all other OW a time-delay and a damping of microbial contamination can be observed. During the flood event TCC in the river Ergolz exceeded 20,000 cells μl^{-1}; with the declining hydrograph they then were reduced until the end of the experiment to approx. 5,000 cells μl^{-1}. Whereas, TCC in the OW were 4 to 6 times lower, they still were considerably high for groundwater samples in general. In contrast, nearly no cells were detected in the regional groundwater flow, represented by the samples taken in OW7.

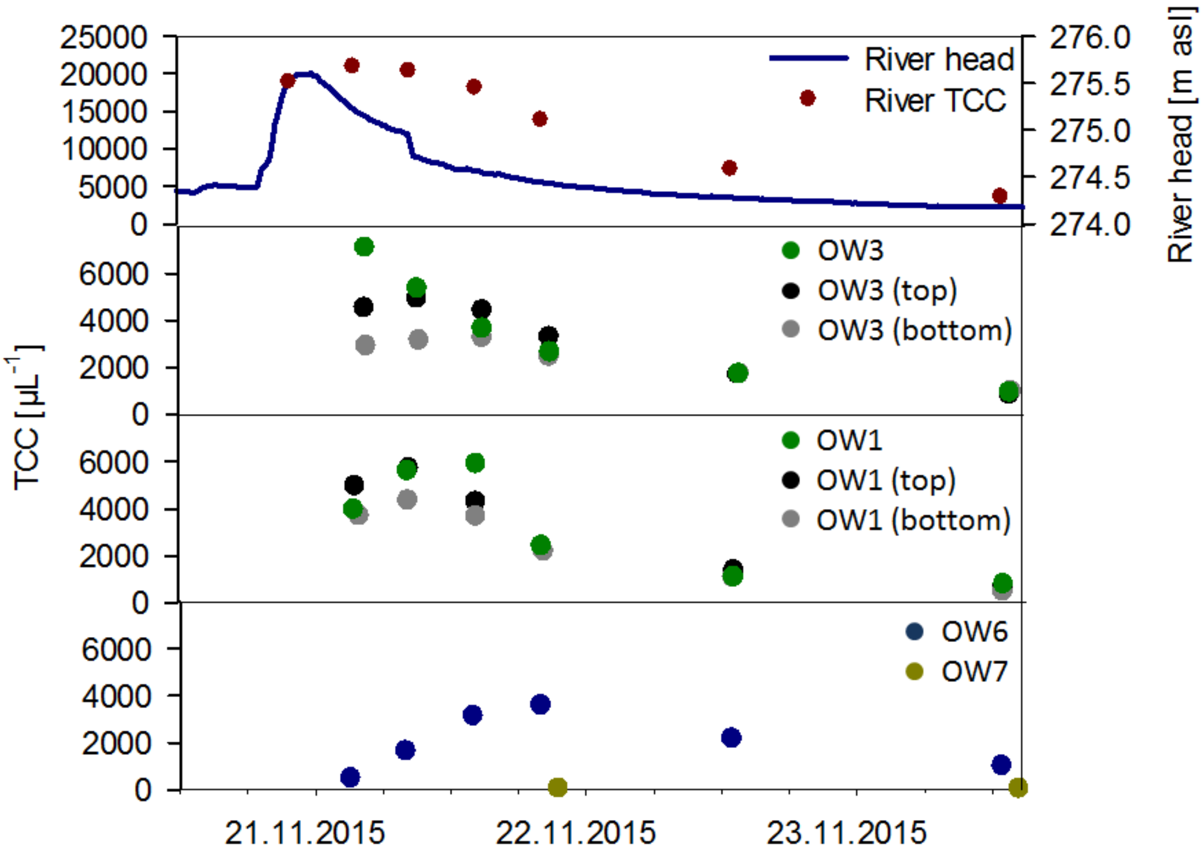

Fig. 7 FCM-TCC data for the flood-event sampling in November 2015 (see Tab. A1) at the investigation area located along the River Ergolz.

The results of the analysed indicator organic MP are summarized in Figure 8 as boxplots for all measurement campaigns performed at the Ergolz GWB. A clear positive correlation of indicator organic MP could be observed between the river and riverine groundwater samples as well as during flood events and dry weather conditions. The highest concentrations for 1H-benzotriazole and acesulfame were measured in the river Ergolz and a clear decrease (factor 2-3) to the groundwater samples taken near to the river was observed. Overall, the lowest concentrations of indicator organic MP are observed in the groundwater samples taken at the sampling location OW7 which is located in the regional groundwater flow regime. At this location the groundwater signature was significantly different compared to all other sampling locations. Carbamazepine, candesartan, lamotrigine, and hydrochlorothiazide were detectable in all groundwater samples. However, in the river water and groundwater samples taken at the sampling location OW7 lower concentrations were observed. Since all above mentioned substances are known to be persistent (Tab. 2), this finding indicates a spatially and temporally displaced earlier entry of river water into the GWB. Atenolol was only detected in the river Ergolz, but the transformation product atenolol acid was still partly detected in the riverine groundwater samples. The persistent pesticides atrazine and terbuthylazine and the transformation product of metolachlor, metolachlor ESA (Tab. 2), were detectable in very small traces in the range of the LOQ. However, no significant differences between the sampling locations were observed regarding these compounds. Therefore it is concluded that for these substances, a basic load is present in the Ergolz GWB, which is only very slowly washed out or diluted. As a result of the baseline sampling performed in December 2013 at the location of the PW, which is located 1.5 km downstream of the investigated river section (Fig. 8), the concentrations of indicator organic MP are generally lower than in the groundwater samples taken close to the river.

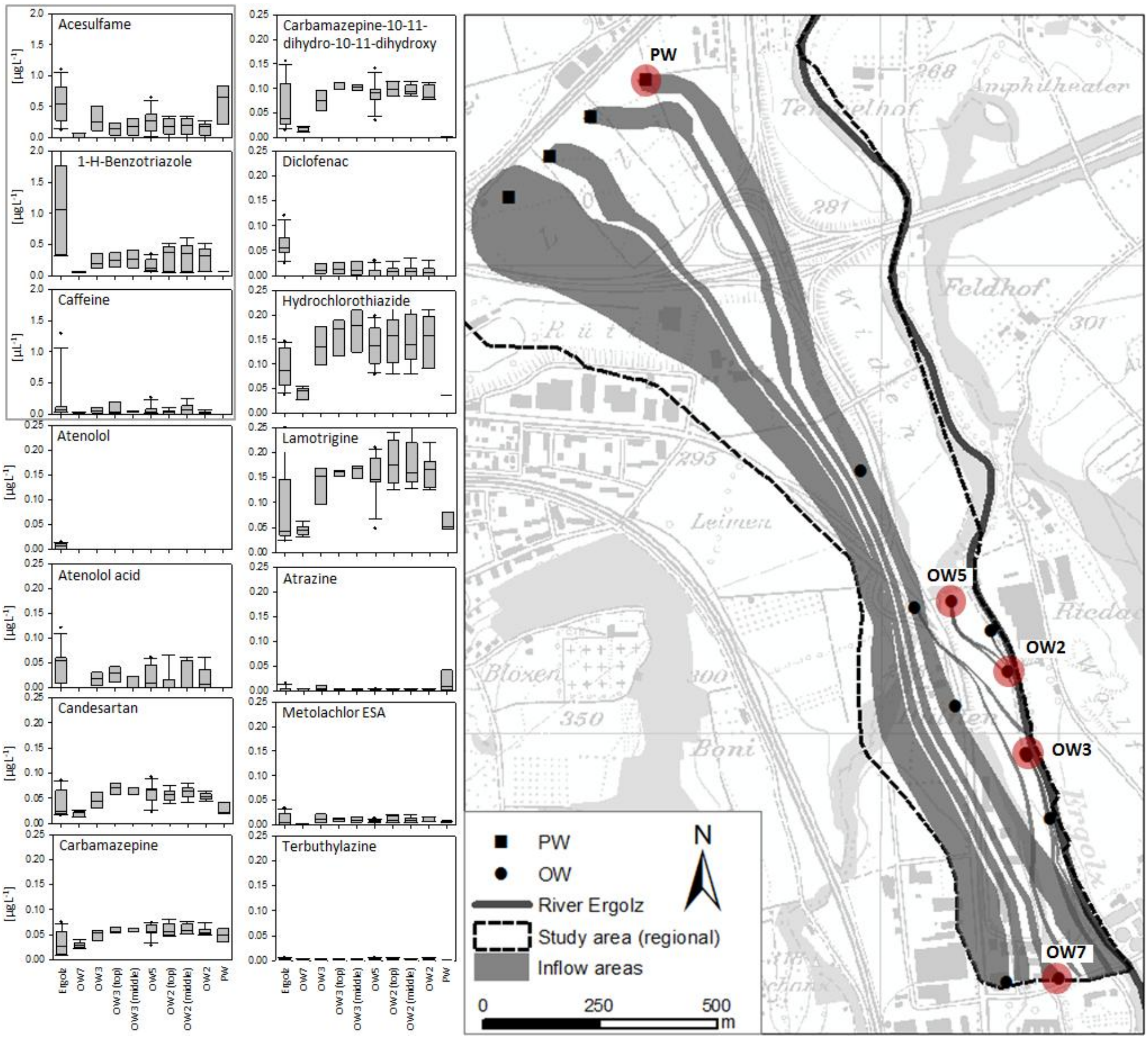

Fig. 8 Left: Descriptive statistics of organic indicator MP (Tab. 2) measured during all campaigns in the Ergolz GWB (gray box shows tracers with concentration resolution up to 2 µgL^{-1}, all others up to 0.25 µgL^{-1}). Right: Sampling locations (red points) related to the regional groundwater flow regime and within the inflow areas of the PW and OW (different hydraulic boundary conditions). Red points: Sampling locations.

Frenke GWB

Figure 9A to E illustrates the hydraulic settings of the Frenke GWB by means of measured river and groundwater heads in relation to the local elevation of the river bed for three OW close to the river Frenke and one OW4 located within the regional groundwater flow system. OW1 and 3 are located in the northern and southern part of the investigation area where the riverine groundwater table was generally below the river bed for the entire measurement period. For these locations, continuous river water infiltration into the unsaturated zone and percolation into the groundwater saturated zone is the dominant process (disconnected interaction type; see Tab. 1). The absolute variations of the hydraulic heads measured in OW4 (Fig. 9E), which is located within the regional groundwater flow system, are higher than those observed in the river (Fig. 9A) indicating that during flood events the GWB is hydraulically controlled by the regional groundwater flow system.

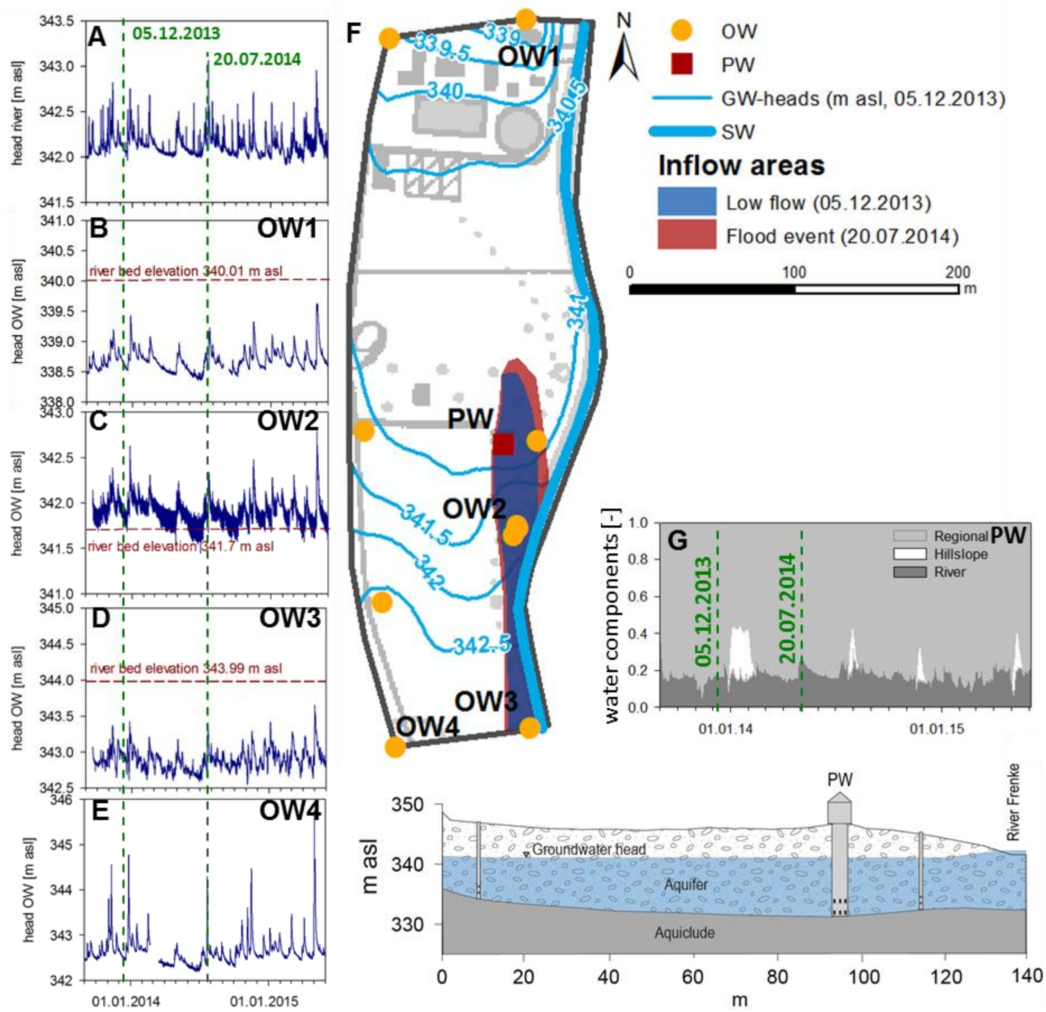

Fig. 9 Groundwater flow regime for the study area located along the River Frenke (Fig. 1). A-E: Measured river and groundwater heads in relation to the local elevation of the river bed for the three riverine OW and one OW located within the regional groundwater flow system. F: Calculated groundwater heads and inflow areas to the PW for a hydraulic situation with low river discharge (05.12.2013) and for a moderate flood event (20.07.2014). G: Calculated groundwater components for the PW. H: Hydrogeological cross section through the aquifer from the river board to the different OW and the PW. Dotted vertical green lines mark the sampled hydraulic situations.

Figure 9F illustrates the variability of inflow areas (filter paths) and the different infiltrating sections along the river during a hydraulic situation with low river discharge (0.8 m^3s^{-1}, 05.12.2013) as well as during a moderate flood event (27.8 m^3s^{-1}; 20.07.2014). During both hydraulic settings the inflow areas to the PW orient towards the river and the regional groundwater flow system. As a result of the influence of the regional groundwater flow on the water composition at the PW during and after flood events, the inflow areas were only slightly wider compared to those during average hydraulic conditions.

Figure 9G shows the calculated water components that constitute the raw water of the PW. During average hydrological conditions the raw water of the PW is constituted by approx. 80% of groundwater components which can be related to regional groundwater flow system; only approx. 15% can be related to infiltrated river water with short residence times (hours to days) and approx. 5% of groundwater components can be related to regional groundwater from the hillslope. During several flood events the hydraulic head of the regional groundwater flow system results in situations where groundwater components, which are related to the regional groundwater flow system from the hillslope, can make up to approx. 30%.

Figure 10 summarizes the results of FCM and fecal indicator bacteria data during a moderate flood event in the river Frenke and the field experiment in July 2014. A clear reduction of microbial contamination from the river to the riverine groundwater and the PW can be observed. Within the first 10 m of the flow paths a 1 log-reduction of TCC and a 2 log-reduction of *E. coli* concentrations can be observed. Against expectation the samples taken in OW4, which is located within the regional groundwater flow system, show higher microbial contamination when compared to those samples of OW3 (location where groundwater infiltration is the dominating interaction type). At this location also the highest concentrations of nitrate was observed. We hypothesize that at the location of OW4 we were able to capture the interaction of the regional groundwater flow system with several fault and the regional Karst systems during flood events. Thereby, bacterial contaminations, which originate from agricultural activities within the catchment area, enter into the alluvial valley aquifer.

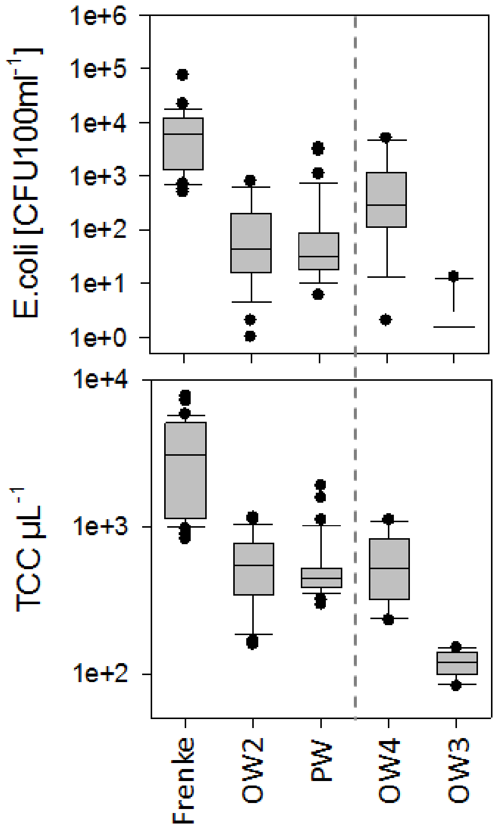

Fig. 10 Illustration of *E.coli* and FCM-TCC data for the flood-event sampling in June 2014 (Tab. A1) at the investigation area located along the River Frenke. The sampling points left of the dotted gray line depict the microbiological data for the hydraulic situation from the River Frenke to the riverine OW2 and the PW; right of the dotted gray line the microbiological data for the two OW related to the regional groundwater flow regime are depicted.

In Figure 11 the results of the indicator organic MP are summarized as boxplots for all measurement campaigns performed at the Frenke GWB. Acesulfame, which can be used as a persistent indicator of waste water entry into rivers and the GWB, shows large variations during flood events and dry weather conditions (Epting et al., 2017). The persistent pesticide metabolite metolachlor ESA as well as the persistent drugs carbamazepine and its transformation product, hydrochlorothiazide and lamotrigine (Tab. 2) were all found in detectable concentrations in all groundwater samples taken close the river. The biologically degradable drugs diclofenac and atenolol (Tab. 2), on the other hand, are much lower in concentration. These relationships confirm that a transformation of biodegradable substances in the hyporheic zone occurs very quickly. Concentrations of 1H-benzotriazole are very heterogeneously distributed within the river and groundwater samples. A non-stationary point source input of industrial origin additional to waste water entries is probable. However, there is no information about the entry location and input function.

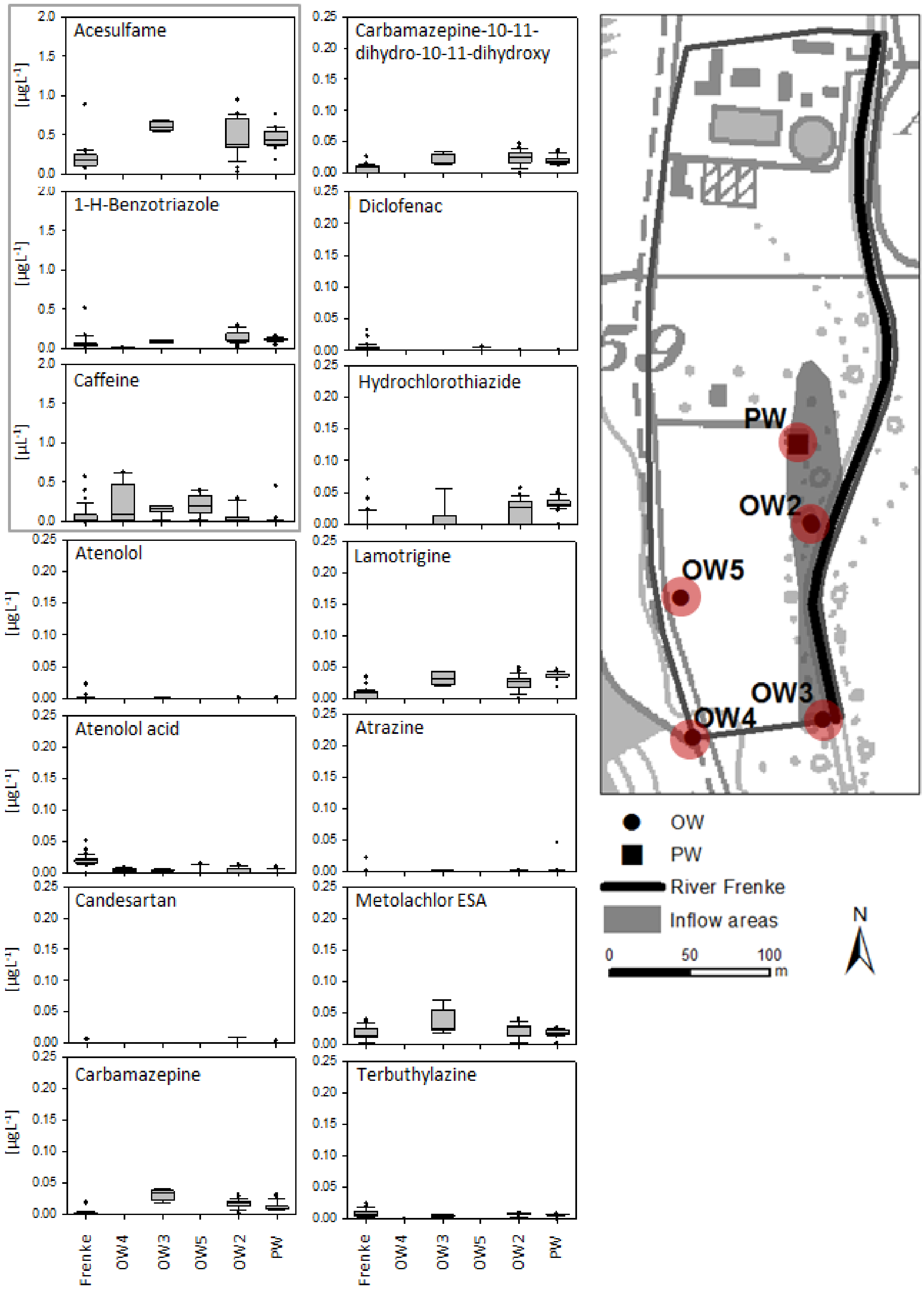

Fig. 11 Left: Descriptive statistics of organic indicator MP (Tab. 2) measured during all campaigns in the Frenke GWB (Gray box shows tracers with concentration resolution up to 2 µgL^{-1}, all others up to 0.25 µgL^{-1}). Right: Sampling locations (red points) related to the regional groundwater flow regime and within the in-flow areas of the PW.

Likewise, tetrachlorethene was detected in low concentrations in PW1 located in the Birs GWB and in the groundwater samples taken close to the river, but not so in the river water itself. Also further to the south, tetrachlorethene was only detected in PW2 but not in the groundwater samples taken close to the river. Similarly tetrachlorethene was detected only in the PW located in the Ergolz GWB. In the Ergolz river water sample and in all groundwater samples in the Ergolz GWB slight traces of diethyl ether were detected, but not so in the PW. In one groundwater sample additionally methyl tert-butyl ether was detected. All other measured VOC were below the detection limit in all three GWB. In the GC-MS screening, in addition to tetrachlorethene, no or only slight traces of natural substances, substances of anthropogenic or industrial origin were detected in the samples including those from the drinking water wells, except for PW1, located in the Birs GWB (Tabs. 4 and 5). However, the origin of the detected substances cannot be conclusively clarified by the investigation carried out but hints to industrial sources and not discharge from wastewater treatment plants.

Table 4 Summary of the positive results of the VOC analysis by means of GC-MS (dry weather conditions in December 2013). Locations and setup of OW clusters and PW: Birs GWB (Figs. 3-5); Ergolz GWB (Figs. 6 and 8) and Frenke GWB (Figs. 9 and 11).

Sample name	Tetrachlorethene µgL^{-1}	Diethyl ether µgL^{-1}	Methyl tert-butyl ether µgL^{-1}
River Birs	<0.05	<0.05	<0.05
OW1 cluster (top)	**1.1**	<0.05	<0.05
OW1 cluster (bottom)	**1.7**	<0.05	<0.05
PW1	**2.9**	<0.05	<0.05
PW2	**0.14**	<0.05	<0.05
OW2 cluster (bottom)	<0.05	<0.05	<0.05
OW2 cluster (top)	<0.05	<0.05	<0.05
River Ergolz	<0.05	**0.14**	<0.05
OW3	<0.05	**0.096**	<0.05
PW	**0.12**	<0.05	<0.05
OW8	<0.05	**0.096**	**0.42**
OW5	<0.05	**0.089**	<0.05
River Frenke	<0.05	<0.05	<0.05
PW	<0.05	<0.05	<0.05

Table 5 Summary of the positive results of the GC-MS screenings (dry weather conditions in December 2013).

	anthropogenic origin	industrial substances	natural substances	unknown
Number of positive results	14	8	4	18
GWB Birs total	8	6	2	4
GWB Ergolz total	6	2	0	9
GWB Frenke total	0	0	2	5

5. Discussion

Context to Regional Groundwater Flow Systems

To facilitate a comparison among the different investigation sites an overall water budget across the main model boundaries of the three study sites as well as a quantification of in- and exfiltration budgets across individual river sections is presented in the following (Fig. 12A-C).

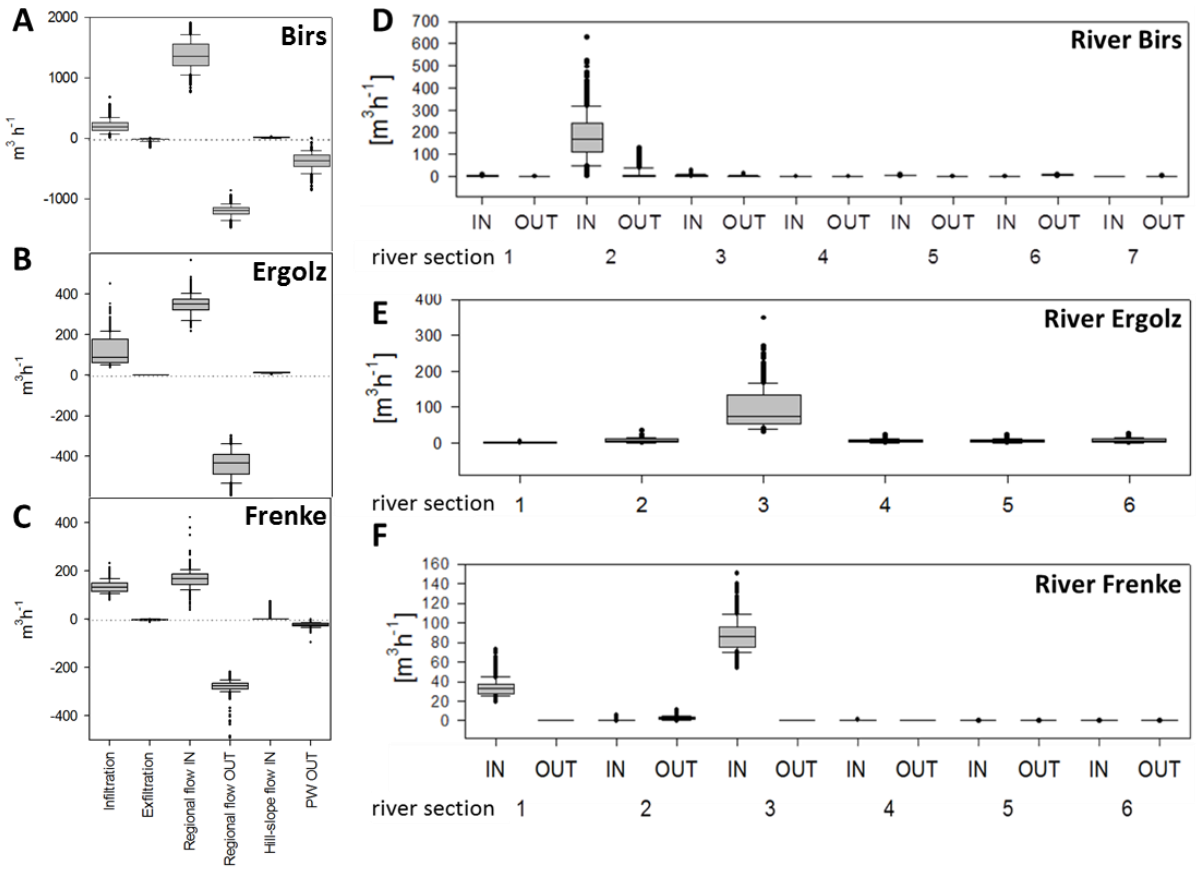

Fig. 12 A-C: Flow budgets across model boundaries. D-F: Flow budgets for river sections (Fig. 2).

For all three study sites along the rivers Birs, Ergolz and Frenke the regional groundwater flow is most relevant in terms of quantity. For the study site along the river Frenke, groundwater flow from the hillslope and the interaction with local fault and Karst systems can be sporadically relevant in terms of quantity. For the study site along the river Birs it could be demonstrated that the hydraulics related to the regional groundwater flow system are of major importance for understanding the local interaction processes. Here, hydraulic signals from high discharge events associated with high flow stages and river water infiltration from southern river sections propagate within the GWB with a considerable retardation of more than half a year. High regional groundwater levels can, along individual river stretches, result in local groundwater exfiltration into the river. However, it has to be mentioned that the weather conditions during this study did not cover extreme events, such as droughts or extreme flood events.

Our investigations demonstrate the importance of understanding the spatiotemporal transient character of regional groundwater flow regimes for quantitative and qualitative issues related to river groundwater interaction. Particularly for the presented case studies in Northwest Switzerland which are characterized by the geological settings of the Tabular and Folded Jura regional groundwater flow regimes are quantitatively of major importance. Here, GWB are often divided by bedrock steps which results in complex sequences of river sections, where either processes of river water infiltration or groundwater exfiltration can be observed (Fig. 12D-F). The geological settings correspond to the "river corridor" concept as introduced by Stanford and Ward (1993). During high discharge events these interaction processes along individual river sections can shift, whereas not necessarily river water infiltration has to increase but processes related to regional pressure propagation can result in increased groundwater exfiltration. Additionally, for the GWB within the valleys of the Tabular and Folded Jura the interaction of the unconsolidated gravel aquifers with local fault and Karst systems have to be considered.

Inflow Areas and Delineation of River Sections

A way to approach riverine groundwater extraction issues and river restoration measures is to assess those river sections which are located within the inflow area of drinking water PW for and during different hydrogeological and operational boundary conditions. The modeling results and the evaluation of different river sections for the investigated GWB illustrate that exchange fluxes across the aquifer-river interface appear to be strongly controlled by scale dependent spatial patterns of streambed hydraulic conductivity and streambed topography (Huber et al., 2013). Brunner et al. (2009) investigated spatial and temporal aspects of the transition from connection to disconnection between rivers and groundwater. They showed that the state of connection is a critical variable in the dynamics of infiltration in a non-steady system. Fluctuating groundwater tables also play an important part in connected and disconnected systems, as they affect the flow regime between surface water and groundwater.

River sections can be delineated according to different hydraulic settings, including (1) connection of river and groundwater systems, including river sections with predominantly infiltration or exfiltration, (2) permanent disconnection of river sections from groundwater systems by a variable thick unsaturated zone, as well as (3) transitional systems with connection during flood events (Tab. 1). For all three study sites the interaction type at different river sections (Fig. 2) could be characterized and, as a result of the groundwater flow modeling, the exchange across the riverbed could be quantified. Intensified exchange was observed along individual river sections (Fig. 13). This can be explained by the local hydraulic settings as well as the hydraulic conductivities of the river bed and the aquifer. For the study site along the river Birs and especially for the southern river sections 1 to 5 river water infiltration into the aquifer is the dominating interaction process for the investigated time period. However, for the northern river sections 6 and 7, quantitatively less significant, groundwater exfiltration into the river can be observed. For the study site along the river Ergolz and most pronounced for river section 3 the infiltration of river water into the aquifer is the only interaction process that can be observed for the investigated time period. For the study site along the river Frenke the infiltration of river water into the aquifer is the dominating interaction process. However, for river section 2, which is directly located within the inflow area of the PW, groundwater exfiltration into the river is the dominating interaction process. For the northern river section 6, both river water infiltration and groundwater exfiltration can be observed.

Composition of Water Components

The calibrated high-resolution groundwater flow models also facilitated comparing the different spatiotemporal settings in the context of regional groundwater flow regimes. The water component approach presented allows an indication of the composition of the water extracted in the PW. Groundwater components B (young river infiltrate) and C (groundwater related to recharge from adjacent hillslopes) are distinct, groundwater component A (regional groundwater) is not distinct, as this component might also include river water infiltrated further upstream and water derived from up-gradient hillslopes. Regarding microbial issues, specifically considering microbial degradation, the "under-estimated" river water components from further upstream are not relevant. This is not true for many organic MP which are persistent in groundwater systems. In this case, the regional groundwater component must be further considered and the question arises, as to whether the substance infiltrated at locations further upstream, or was released within the regional inflow area or hillslope catchment. To answer this question, further characteristic substances which undoubtedly originate from activities in the inflow area (municipal waste, substances related to brown fields, etc.) must be evaluated and put into relation with the occurrence of certain substances from other sources.

The evaluation of groundwater components of the investigated GWB at the Birs, Ergolz and the Frenke also illustrate that at a larger stream-reach to sub-catchment scale, exchange fluxes between groundwater and surface water is strongly affected by larger geological heterogeneities in the alluvial aquifer and the resulting groundwater flow field (Cardenas and Wilson, 2007; Engdahl et al., 2010; Fleckenstein et al., 2006; Huggenberger et al., 2013)

Microbial Contamination and Filter-efficiency

Peaks observed in microbial TCC during high discharge events primarily can be related to bacterial inputs such as river sediments, surface run-off, agricultural run-off, and wastewater overflows (Frey et al., 2015; Hill et al., 2006). This was shown previously for indicator organisms (Page et al., 2012) and can generally be expected due to the known differences in TCC between surface waters (usually in the order of 10^3 cells μl^{-1}, (Wang et al., 2007)) and groundwater (usually in the order of 10^1 cells μl^{-1}, (Sinreich et al., 2014)). As observed in previous studies related to fecal indicator bacteria and in regard to the water extracted from the PW in the three study sites, the quality standards for drinking water during average flow and surface water quality conditions is satisfied (Regli et al., 2003; Taylor et al., 2004). In detail, for both sampled PW (PW1 and PW2) in the study areas along the river Birs, filter paths from the location of infiltration to the PW are in the order of 370 to 450 m over which the microbial load is reduced by two orders of magnitude, resulting in average filter-efficiencies of approx. 1E-02 und 1.5E-02 m^{-1} (Fig. 13). In comparison, microbial TCC and fecal indicator bacteria concentrations at the study site along the river Ergolz, were one order of magnitude higher. This finding is in accordance with the large fraction of groundwater components that can be related to the infiltration of river water with short residence times and the generally high microbial contamination of surface waters, especially during flood events. However, the filter paths from the location of infiltration to the location of OW5 is in the order of 275 m and microbial contamination is reduced by two orders of magnitude, resulting in average filter-efficiencies of approx. 1.3E-02 und 1.8E-02 m^{-1} (Fig. 13). With this, the calculated average filter-efficiencies of the Ergolz system are higher than those of the study site at the Birs river, although TCC and fecal indicator bacteria concentrations are higher at the Ergolz compared to the Birs.

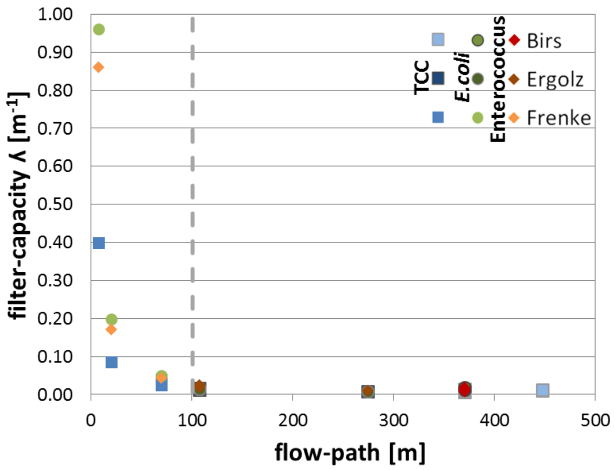

Fig. 13 Compilation of filter-efficiencies (in relation to concentrations measured within the rivers) for the fecal indicator bacteria *E.coli* and Enterococcus as well as TCC derived from flow cytometry for all study sites in dependence to the distance to the river (derived from modeled flow paths to the individual sampling locations). From distances < 100 m a derivation of filter-efficiencies is not distinct as groundwater also domiciles indigene microbial communities and FIB are already removed. Dotted gray line: 100 m flow-path distance.

At the study site along the river Frenke elevated fecal indicator bacteria concentrations of the pumped raw water (Fig. 10) were observed. Here, the filter paths from the location of infiltration to the location of the PW is in the order of only 70 m and microbial contamination is reduced by two orders of magnitude, resulting in average filter-efficiencies of approx. 4.2E-02 und 1.2E-01 m^{-1} (Fig. 13), which are considerably higher than those observed at the study sites along the rivers Birs and Ergolz.

Fate of organic MP and derivation of indicator substances

Organic MP which originate from municipal wastewater systems follow three main input pathways into surface water systems which can be distinguished: (1) treated sewage water from wastewater treatment plants; (2) non-treated sewage water deriving from storm water overflow discharge during capacity exceedance of wastewater treatment plants and sewage systems; and (3) leakage from sewage systems erroneous pipe-connections. Further organic MP can originate from application of pesticides to agricultural land and the spreading of manure which leaches via the soil to the groundwater or through drainage or agricultural run-off to the surface water (Derksen et al., 2004).

For the three investigated GWB along the rivers Birs, Ergolz and Frenke the number and concentrations of organic MP within the surface water and groundwater samples are similar. No significant differences between the study sites were observed. This is a result of the similar activities, including urbanization and agriculture, within the catchment areas of the study sites (Fig. 1). The organic MP concentrations observed for the groundwater samples are in the same range as have been observed for similar studies along the Swiss river Thur (Huntscha et al., 2012; Huntscha et al., 2013) and in average for studies related to contaminations of European groundwaters (Loos et al., 2010). However, compared to other European groundwaters no extreme concentrations have been observed.

Several organic MP were observed, which are only slowly degraded or diluted within the aquifers. Considering all measured values a slight tendency of higher concentrations for the study site along the river Ergolz can be observed. This observation is in accordance with the higher fraction of sewage water of the river Ergolz. As many persistent substances are transported within the regional groundwater flow regime, which also is partly fed by river water that has infiltrated up-gradient of the study site, a differentiation of the different groundwater components on basis of organic MP is difficult. The dynamic organic MP-input-function related to infiltrating river water during flood events could only partially be reproduced for the Frenke GWB (Epting et al., 2017).

In general, during high river discharge events larger fractions of sewage water as well as fertilizers washed out from agricultural land can lead to elevated organic MP concentrations first in the rivers and then later in the groundwater systems. At the same time variable hydrological boundary conditions can lead to reduced residence times of groundwater components within GWB. This is relevant especially for PW close to rivers. As a result often short-term elevated organic MP contaminations can be observed which can be faced by adaptive groundwater management (operational regime) or adequate raw water treatment (e.g., active carbon).

6. Conclusion

Investigations related to groundwater management and river restoration issues often are limited to a local view, e.g. between interacting river sections and nearby drinking water PW. The results of our investigations illustrate the influence of dynamic hydrologic boundary conditions on river-groundwater interaction and of regional scale groundwater flow regimes on the water composition of PW close to rivers.

It could be demonstrated that the applied approach allows identifying river sections and their variations with intensified river-groundwater exchange processes. Furthermore, the transient character of the different groundwater components that constitute the raw water quality of drinking water wells near rivers could be quantified. This knowledge, together with the derived proxy indicators for the diverse contaminations, presents the basic elements for both groundwater management and river restoration concepts. Furthermore:

- Investigations of three individual case studies enabled us to compare the characteristics of different groundwater flow regimes.
- A quantification of groundwater components allowed us to describe how raw water quality is composed for different boundary conditions.
- Local river-groundwater interaction processes are highly relevant for microbiological contaminations, whereas, regional groundwater components play an important role especially for contaminations by organic MP.

- Evaluating individual river sections enabled us to identify river sections that are located within the inflow area of drinking water PW for different spatiotemporal settings.
- This information allows an optimized procedure of restoration measures, whereby river sections with predominantly exfiltrating groundwater conditions or those which are decoupled from the groundwater surface should be preferred.
- We were able to derive filter-efficiencies as a function of aquifer characteristics, distance and hydraulics.
- Our data show that the banks and beds of the investigated rivers act as a primary barrier to many contaminants. The filter-efficiency of the aquifer material further acts as a natural physical, biological, and chemical filter and reduces contaminant loads as water passes through the subsurface matrix.
- The identification of indicators for river water infiltration in line with microbial and organic MP contamination is an important step in managing groundwater resources and hazard assessment.

Risk assessment and management should take into account short- and long term measures, including (A) the optimization of monitoring strategies and operational regimes of drinking water wells close to rivers, and (B) the formulation of remedial measures regarding urban drainage and agricultural activities in the catchment areas.

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Table A.1 Settings of measurements as well as experiments performed at the three study areas, including different monitoring systems, located within the rivers, groundwater OW and PW.

	Birs	Ergolz	Frenke
Experimental sites (Fig. 1)	Two river sections experimental sites A & B	Three river sections experimental site A, B & C	One experimental site
OW* & PW** ***measured parameters***	**8 OWs and 3 clusters a 3 OWs** *h^{+}; T^{++}* *selected with EC^{+++}; pH; FNU^{++++}; SAC^{+++++}; particles 2 – 10 µm; online* **8 PWs** *Extraction rates*	**6 OWs** **2 OWs; 3 CMTs*** *(DirectPush)*** *h^{+}; T^{++}* *selected with EC^{+++}; FNU^{++++}; online*	**5 OWs** **3 OWs; 2 CMT *(DirectPush)*** *h^{+}; T^{++}* *selected with EC^{+++}* **1 PW** *Extraction rates; EC^{+++}; pH; FNU^{++++}; SAC^{+++++}; online*
River measurements	**1 downstream experimental site B** *h^{+}; Q; T^{++}; online* (BAFU 2106) **1 experimental site B** *h^{+}; T^{++}; EC^{+++}; pH; FNU^{++++}*	**1 between experimental sites A & B** *h^{+}; T^{++}; EC^{+++}; online* **1 downstream experimental site C** *h^{+}; T^{++}*	**1 experimental site** *h^{+}; T^{++}; EC^{+++}; online*
Regional geological settings **Groundwater body**	aquifer bottom/aquitard: Tertiary deposits Elsässer Molasse (sandstone marl bedrock) maximal depth to groundwater 29 m; thickness of saturated zone 0.6 – 10 m.	aquifer bottom/aquitard: Tertiary deposits Meletta layers (clay-/sandstone bedrock) maximal depth to groundwater 41 m; thickness of saturated zone 0 - 12 m.	aquifer bottom/aquitard: Jurassic deposits Hauptrogenstein (carbonates) and Unterer Dogger (layering of marls, claystones and carbonates); interaction with karstified Jurassic carbonate formations (Hauptrogenstein, carbonate rock) maximal depth to groundwater 8.2 m; thickness saturated zone 8.8 – 14.2 m
Experiments ***Time period (boundary conditions)***	**Routine measurements[1]** *20.12.1999 - 19.01.2015 (diverse)* **Specific measurements** *09.06.2009 - 09.05.2010 (diverse)[2]* *24.07.2012 - 25.07.2012 (test)[3]* *02.12.2013 - 05.12.2013 (dry)[3]* *01.01.2015 - 01.04.2015 (diverse)[3]* *05.04.2015 - 21.04.2015 (minor flood event)[3]*	**Specific measurements** *20.07.2009 - 26.08.2009 (diverse)[2]* *28.09.2011 - 25.06.2012 (diverse)* *02.12.2013 - 05.12.2013 (dry)[3]* *10.07.2013 - 29.10.2014 (diverse)[3]* *20.11.2015 -23.11.2015 (flood event)[3]*	**Routine measurements[1]** *20.12.1999 - 19.01.2015 (diverse)* **Specific measurements** *02.12.2013 - 05.12.2013 (dry)[3]* *17.03.2014 (dry)[3]* *15.05.2014 – 22.05.2014 (test)[3]* *20.07.2014 – 26.07.2014 (flood event)[3]* *06.10.2014 – 22.10.2014 (flood event)[3]* *08.10.2013 - 29.10.2014 (diverse)[3]*
Hydrogeophysical experiments ***detected features***	ERT' along river board *bedrock; zones preferential R-GW-I'''* GPR'' inflow area of PW *aquifer layering; dimension depositions*	ERT' along river board *bedrock; zones preferential R-GW-I'''*	ERT along river board and hillslope *bedrock; zones preferential R-GW-I''*; fracture structures

OW*: Observation Well; PW**: Pumping Well; CMT***: Continuous Multichannel Tubing; *h^{+}: hydraulic head; T^{++}: Temperature; EC^{+++}: Electrical Conductivity; FNU^{++++}: Formazine Nephleometric Unit; SAC^{+++++}: Spectral Absorption Coefficient (at 254 nm); Q: Discharge;* ERT'= Electrical Resistivity Tomography; GPR''= Ground Penetrating Radar; R-GW-I'''=River-GroundWater-Interaction; [1]performed by the Cantonal Laboratory Basel-Landscape; [2]Polat-Elma (2010); [3]project "regional water supply Basel-Landschaft 21"

Table A.2 Software, model geometries, parameterization and calibration.

	Birs	Ergolz	Frenke
software	GMS 10.0.11 (Finite Differences)		
codes	MODFLOW 2000 (GW-Flow; (Harbaugh et al., 2000)) MT3DMS (GW-Transport; (Zheng and Wang, 1998)) PEST und Pilot Points, model calibration (Doherty, 1994; Doherty, 2003)		
resolution [m] (number of model cells [#]) (number of layers [#])	10 6682 1	10 4018 1	10 637 1
model geometries			
perimeter	Delineation lower terrace gravels, main groundwater current		
bedrock surface topography	GeORG-project (www.geopotenziale.eu; (Dresmann et al., 2015)) adaptation on basis of DPT-probes and hydrogeophysics DHM25 and DHM2m (SwissTopo)		
river bed	survey TBA (digital): longitudinal and transverse profiles (recording 20.08.1946 to 13.01.2006)	survey TBA (analogue) longitudinal profiles Ergolz (1:2000 / 200), WK – 29; municipalities Füllingsdorf / Pratteln / Augst (recording December 1978)	survey TBA (analogue) longitudinal profiles municipalities Liestal, Seltisberg, Bubendorf WK 95a (date 30.04.2010)
model parametrization			
time resolution	1h		
regional groundwater current from South to North	Dirichlet, head (continuous GW-head-measurements; partially interpolation GW- head-measurements to model boundaries)		
regional hillslope to the West	Neumann, flux (estimated on basis of the subsurface catchment to the model boundaries and annual precipitation amounts, minus evapotranspiration; differentiation between sealed and non-sealed areas)		
surface water	Cauchy, head and conductance*		
drinking and process water supply	Neumann, flux (conditioned data)		
GW-renewal	Neumann, flux (estimated on basis of annual precipitation amounts, minus evapotranspiration; differentiation between sealed and non-sealed areas)		
results model calibration			
hydraulic conductivity; k_f-values [ms^{-1}]; IDW**-Interpolation between Pilot Points	1.5E-05 – 9.7E-02	1.1E-03 – 1.0E-02	1.4E-07 – 1.4E+00
horizontal anisotropy [-]	1.4E-01 – 7.8E+00	2.5E-01 – 1.0E01	6.3E-04 – 1.0E02
conductance OW [ms^{-1}]; river sections	9.3E-07 – 8.9E-04	3.3E-06 – 3.7E-04	9.6E-07 – 9.4E-04
Sy*** Ss**** n*****		0.08 (defined) 1.0E-4 (defined) 0.08 (defined)	

*river bed conductance; **Inverse Distance Weighted; ***Specific Yield; ****Specific Storage; *****Porosity