Science of the Total Environment 599–600 (2017) 227–236

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Online analysis: Deeper insights into water quality dynamics in spring water

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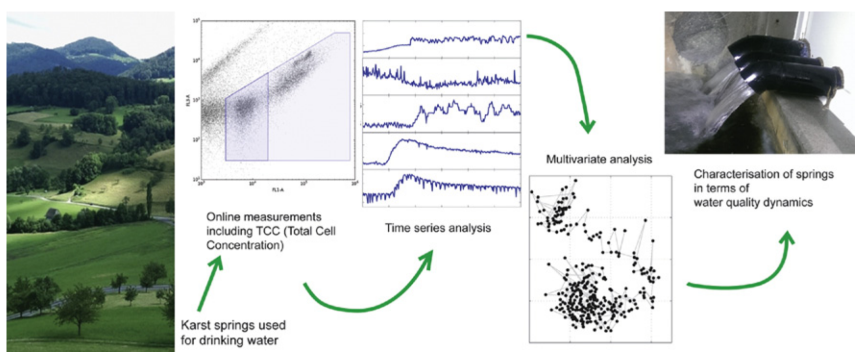
HIGHLIGHT

- Karst spring water quality mostly has a high spatio-temporal heterogeneity.
- Total Cell Count (TCC) in spring was sensitive to precipitation in catchment area.
- High-resolution monitoring of TCC and abiotic parameters elucidated event-driven dynamics.
- Non-linear relationship between TCC and abiotic parameters (incl. SAC254 and turbidity)
- Neural-network based multivariate time-series analysis approximated TCC dynamics

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 27 February 2017
Received in revised form 26 April 2017
Accepted 27 April 2017
Available online 3 May 2017

Editor: D. Barcelo

Keywords:
Karst spring water
Drinking water
Online monitoring
Online flow cytometry
Multivariate time series analysis

ABSTRACT

We have studied the dynamics of water quality in three karst springs taking advantage of new technological developments that enable high-resolution measurements of bacterial load (total cell concentration: TCC) as well as online measurements of abiotic parameters. We developed a novel data analysis approach, using self-organizing maps and non-linear projection methods, to approximate the TCC dynamics using the multivariate data sets of abiotic parameter time-series, thus providing a method that could be implemented in an online water quality management system for water suppliers. The (TCC) data, obtained over several months, provided a good basis to study the microbiological dynamics in detail. Alongside the TCC measurements, online abiotic parameter time-series, including spring discharge, turbidity, spectral absorption coefficient at 254 nm (SAC254) and electrical conductivity, were obtained. High-density sampling over an extended period of time, i.e. every 45 min for 3 months, allowed a detailed analysis of the dynamics in karst spring water quality. Substantial increases in both the TCC and the abiotic parameters followed precipitation events in the catchment area. Differences between the parameter fluctuations were only apparent when analyzed at a high temporal scale. Spring discharge was always the first to react to precipitation events in the catchment area. Lag times between the onset of precipitation and a change in discharge varied between 0.2 and 6.7 h, depending on the spring and event. TCC mostly reacted second or approximately concurrent with turbidity and SAC254, whereby the fastest observed reaction in

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http://dx.doi.org/10.1016/j.scitotenv.2017.04.204

the TCC time series occurred after 2.3 h. The methodological approach described here enables a better understanding of bacterial dynamics in karst springs, which can be used to estimate risks and management options to avoid contamination of the drinking water.

1. Introduction

Karst aquifers are an important source of drinking water serving around 25% of the world's population (Ford and Williams, 2007) and regionally can be the dominant source of drinking water. The characteristic, highly soluble bedrock of karst systems (e.g., carbonate, gypsum) is dissolved over time, leading to continuously enlarging water-bearing networks (Quinlan et al., 1992). Transport through these networks is defined by two flow components: (1) rapid flow of groundwater through conduits and joints and (2) diffuse flow through the limestone matrix (Butscher et al., 2011). The rapid flow of groundwater results in a high temporal heterogeneity (i.e. stronger, more rapid, and more frequent variations) of spring water quality, especially after precipitation events (Field and Nash, 1997). This can lead to much lower attenuation of substances in the karst network, in particular of microbiological pollutants (Murray et al., 1981). The often unknown spatial extent and structure of the conduit network of karst aquifers makes the delineation of surface and subsurface catchment areas difficult and thus hinders watershed protection. Given the outlined complexity and in particular the temporal and spatial variability, ensuring high drinking-water quality is challenging in karst areas. The structural complexity of karst systems (e.g., series of reservoirs and interactions between geological compartments) can modify the response of discharge to precipitation events in different springs (e.g., varying lag time between precipitation and increase in discharge) (Taylor and Greene, 2008).

Drinking water supply systems in karstic areas in Switzerland currently mostly record the amount of water entering their system (e.g., spring discharge or other sources) and turbidity. The information on the quantity has two purposes: (1) financial: how much drinking water was produced and how much revenue was generated, (2) demand: if an insufficient quantity of water is produced, alternative sources need to be sought to ensure sufficient supply to customers (mostly buy-ins from associated drinking water supply systems). The turbidity measurements are used as a trigger to automatically discard the raw water if the threshold value for effective UV-disinfection (usually 0.5 FNU) is exceeded, but are also occasionally applied in water utilities without UV-disinfection. In contrast to the online monitoring of these two abiotic operational parameters, microbiological water quality monitoring is based on infrequent grab samples and cultivation-based laboratory analyses of heterotrophic plate counts and indicator bacteria (e.g., *E. coli*, *Enterococcus sp.*). The sampling frequency is defined based on the number of customers and can be as low as two to four controls per year, but can be increased if problems were detected. This is of concern, because standard protection approaches of catchment areas are hard to apply in karstic areas. Furthermore many water utilities use only a UV-disinfection and some do not treat their water at all. Consequently, water quality monitoring at appropriate temporal resolution is particularly important in karst springs to better understand resulting risks and manage them appropriately (Quinlan et al., 1992; Ryan and Meiman, 1996; Auckenthaler et al., 2002). This is not achieved by infrequent monitoring as short-term dynamics are not captured.

Over the years, technological advancements have made it feasible to measure a number of physical and chemical characteristics beyond the above-mentioned operational parameters at high temporal resolution and online (Pu et al., 2011). This has resulted in increased physicochemical monitoring studies (Trcek, 2008; Mudarra et al., 2012) often in combination with modeling (Mohammadi and Field, 2009; Ghasemizadeh et al., 2012). Given the increased risk for microbiological contamination during changing conditions in (karst) springs (e.g., after precipitation events), a number of studies have also tried to link observed dynamics in abiotic parameters with fecal contamination as a major water quality risk: They explored the potential of more easily measureable abiotic parameters such as particle counts and the spectral absorption coefficient at 254 nm (SAC254) as indicators for the presence of organic matter (Shutova et al., 2014) and of microbiological indicator organisms (Pronk et al., 2007; Stadler et al., 2010). Attempts to go beyond direct comparison between abiotic parameters and microbiological contaminations include multivariate analysis methods that can aggregate dynamics in multiple parameters into a measure of change in overall system state (Page et al., 2015).

The current lack of direct microbiological measurements with sufficiently high temporal resolution, resulting from technical limitations in automation of cultivation-based detection methods, limits the ability to investigate the biotic and abiotic factors contributing to dynamically changing water quality. Strategies to increase knowledge on temporal microbiological dynamics include targeted samplings with autosampling devices (Stadler et al., 2008) as well as new and rapid, online detection methods for example using enzymatic activity as a proxy for bacteria (Ryzinska-Paier et al., 2014). Recently, fully automated, insitu measuring systems for high-frequency measurements of total cell concentration (TCC) based on flow cytometry were presented (Brognaux et al., 2013; Besmer et al., 2014).

This study investigates karst system-dynamics linked to precipitation-based events. The analysis is based on the longest high-frequency, multivariate abiotic and online microbiology data set to date. The combination of meteorological, microbiological and physicochemical (i.e. abiotic) parameters from three separate springs located in one study area provides a basis to study the system-dynamics without having to account for differences due to different geographical settings. This study combines high-resolution time series of microbiological and abiotic parameters by neural network-based analysis to address a gap between existing and new monitoring approaches to drinking water quality. With the broader goal of improving the understanding of karst systems and their temporal dynamics, the specific aims of this study were to: (1) identify system-dynamics based on microbiological and abiotic parameters following precipitation events, (2) elucidate and compare the effect of different precipitation events on different springs in the same region and (3) test the multivariate integration of the abiotic parameters to describe dynamics as changes in system state.

2. Materials and methods

2.1. Study site and measurement setup

The study was located in north-western Switzerland, where the folded Jura encounters the tabular Jura, a low mountain range composed of predominantly flat-lying Jurassic and Triassic sediments. The site encompassed a karst system within the imbrication zone (flat and ramp structures overthrusting the tabular Jura). The hydrogeological context of the investigated area is shown in Fig. 1 and information on the larger area is described in Matousek (1985).

Three karst springs were included in the study, all of which occur at the stratigraphic contact between highly karstified limestone (Hauptrogenstein and Trigonodusdolomit) representing the aquifer and the underlying Anhydrite zone representing the aquiclude. The three springs are located close to each other so that precipitation in this region would affect all of them. Despite the proximity of the springs,