KARMA

Karst **A**quifer **R**esources availability and quality in the **M**editerranean **A**rea

Preparatory actions for the development and implementation of Early Warning Systems (EWS) for spring water contamination

Deliverable 3.2

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Executive Summary

This second deliverable of WP3 (Water Quality) deals with the preliminary actions performed to design and develop Early Warning Systems (EWS) for the detection of spring water contamination episodes in selected water supplies across the Mediterranean area. At individual test sites, the main characteristics of the karst aquifers and groundwater supplies, the water quality issues to justify the implementation of an EWS, its typology and monitoring set-up, the first results, the data management and modeling strategy and the expected operational benefits are presented. Precedent hydrogeological studies and existing mid/long-term monitoring records have been used to better characterize the hydrogeological systems studied, which are useful to establish a suitable sampling/data record strategy. The groundwater monitoring network in all case studies has common research approaches by which similar methods and field devices set-ups have been applied. These are: fluorescence-based techniques, particle size distribution (PSD) and microbiological and culture-based methods (i.e. *E. Coli*), which are complemented with further measurements/determinations of specific chemical/isotopic/microbiological parameters (e.g. dye tracers, water stable isotopes, ^{222}Rn, DNA analysis, etc.). This combination of hydrogeological data will allow an effective tracking of groundwater flowpaths from the contamination sources to the captured springs. All study cases have shown similar groundwater pollution-related issues, of which urban waste waters is of a major human concern, but also all those contaminant sources related to the agriculture and cattle-related activities. These common water contamination problems will ensure the replicability of the methods applied worldwide under similar circumstances in karst terrains. In this report, a wide variety of new and existing hydrographs and chemographs have been presented for the four selected sites, showing the first promising results and the establishment of the well-shaped monitoring networks. Most of the preliminary outcomes are orientated to understand transport processes (particles, solutes, bacteria, etc.) and the potential degradation of contaminant substances throughout karst connections. In a more advanced step, adapted to site-specific and operational characteristics, refined protocols should be performed facing the implementation of individual EWS. The remote data gathering via telemetry systems and its integration with smart algorithms for warning messages remain challenging to achieve the most advanced technological level and the potential automatization of operations works from the raw water capture facility. This will greatly contribute to effectively manage and protect karst groundwater resources, but also to minimize human risk against accidental/prolonged intake of polluted groundwater.

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1. Introduction

Since last decades, contamination events in (surface) drinking water sources have been widely studied and scientists and water managers have jointly designed and applied different methods to face this issue. Early Warning Systems (EWS) have emerged as a promising tool to detect the arrival of polluted water in drinking water sources, but to date they have been little implemented in water supply systems partially or entirely fed by groundwater. Among hydrogeological systems by which groundwater flows, karst aquifers very often show certain similarities with surface water schemes (hierarchized drainage, fast flow dynamic, high vulnerability, etc.) that make them ideal for EWS development.

The basic idea of an EWS for karst spring water contamination detection is to identify parameters, or a set of specific parameters, that are easy to monitor (such as turbidity, organic carbon or electrical conductivity) and that indicate the presence of different contaminants (e.g. faecal bacteria, pesticides, antibiotics, toxic metals) (Pronk et al. 2007). For that purpose, different approaches can be adopted depending on the groundwater monitoring network, which needs to be adapted to site-specific conditions and water quality issues in urban water supplies.

In the three KARMA test sites and in the additional test site in the Austrian Alps (which was used for testing and validating the used methods and parameters; Fig 1.1), where EWS are being developed, specific parameters and methods are monitored/applied to understand the occurrence of contamination events at different time-scales:

- fluorescence-based techniques, which allow to record pollution indicators such as turbidity, NOM or tryptophan;
- particle size distribution (PSD), for a better comprehension of the physical transport processes;
- microbiological and culture-based methods, which allow to detect and quantify the presence of pathogen organisms (total coliforms, *E. Coli*, etc.);
- trace metals analysis, which may threaten human health when exposed to high concentration for long periods and can be associated with leakage of urban waste waters (e.g. Gd); and
- ^{222}Rn gas measurements, which can be used to trace the karst flows.

Figure 1.1. Selected KARMA test sites for Early Warning System development and implementation.

In this report, a full overview of individual EWS developed in different test sites is presented, aiming to predict contamination events and minimize the effects of polluted karst groundwater in drinking water supplies across the Mediterranean area.

2 The Qachqouch aquifer (Case Study Lebanon)

2.1 Characteristics of the karst groundwater supply

The Qachqouch karst spring (Lebanon) drains a catchment area estimated at 56 km^2 consisting of limestone and dolostone of Jurassic age (Dubois et al., 2020). It has a yearly volume ranging between 40-60 Mm^3, with maxima reaching 17 m^3/s during flood times. Its flow rate is about 300 l/s during baseflow (July-September). Springwater is discarded into the adjacent El Kalb River during high flow periods (December- May) and utilized during the low flow periods (May-December) to supply northern Beirut while other water resources (adjacent Jeita spring and deep Qachqouch wells) face a deficit. Its annual mean discharge reaches 2 m^3/s. The water is channeled through a canal to a treatment station in Dbayeh Area 5 km downstream where conventional chlorination takes place before the water is supplied into water conduits to northern Beirut. Along with other resources, it contributes with 1-5 m^3/s in total to the water supply of 1.5 million inhabitants in the capital Beirut (MoEW). In the case of high apparent turbidity events (estimated qualitatively >50 NTU; Figure), the water is not supplied in the water network. During high flow, the water level is observed to increase to exceptional levels (above 3 m) inside the spring channel, which requires intervention to avoid flooding inside the spring capture zone.

a b

Figure 2.1. a) Qachqouch Spring during high flow; b) the El Kalb River that infiltrates into the Qachqouch spring (High turbidity observed in January 2016).

2.2 Sources of Pollution: risks and hazards

Its rural catchment encloses about 6% urban area, with 23% forests, 46% grassland and scrubland, 15% cultivated trees (vineyards, olive, and fruit trees), and 10% varied field crops (Figure 2.2). The major source of infiltration, which may lead to the infiltration of contaminants are as follows:

1) point-source infiltration

a) Sinking stream from a heavily polluted river (El Kalb River; 3% of infiltration)

b) Dolines located in the upstream catchment area

c) Untreated wastewater bottomless pits or abandoned wells

2) Diffuse infiltration through cultivated lands

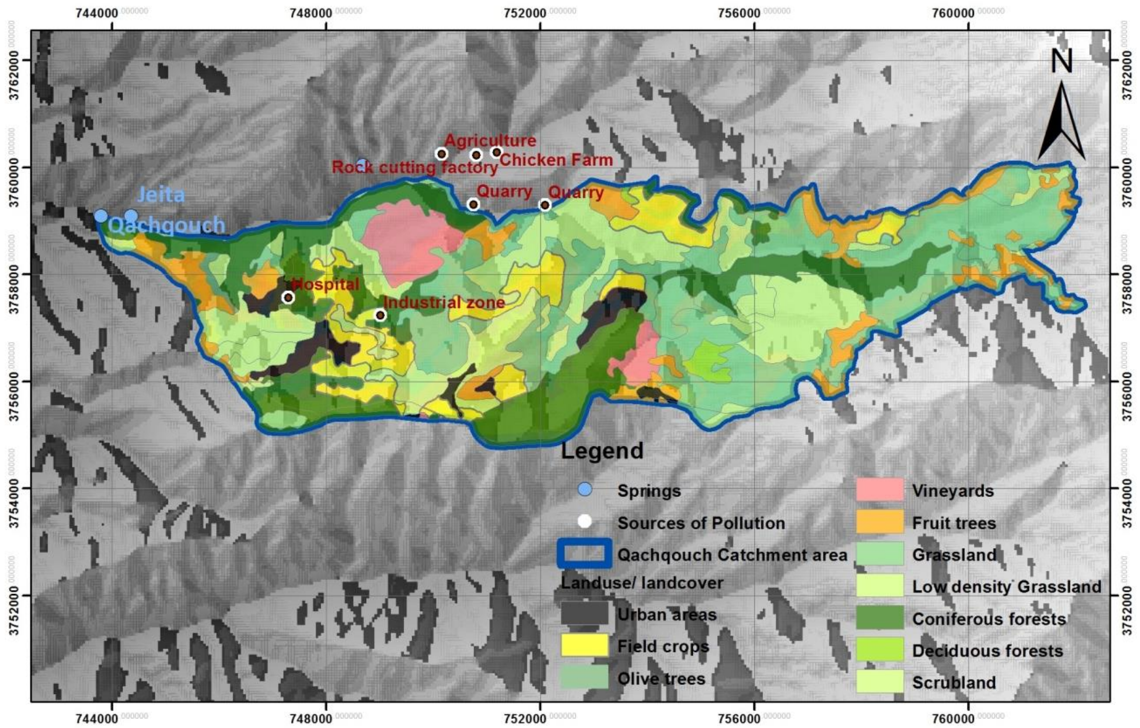

Figure 2.2. Land use and land cover in the Qachqouch catchment and potential recorded sources of pollution

The main sources of hazards on the catchment consist of domestic untreated sewage directly discharged into the environment and the River, hospital waste from selected private hospitals, industrial waste from quarries (fine suspended particles and lubricants; Figure 2). The agricultural waste consists of manure and pesticides spread diffusively in selected cultivated areas. Potential heavy metals or oil products and by-products could be generated from fuel stations found on the catchment.

The types of contaminants reported in the spring are mostly 1) bacteria (total coliform, *E. coli, streptococcus*), as well as 2) suspended particles (Doummar and Aoun, 2018a, 2018b, Allouche, 2020). Other micropollutants indicative of hospital and agricultural waste, as well as pharmaceuticals, are found in concentrations below toxic levels even during contaminant breakthrough (Doummar and Aoun, 2018b). Other types of oil-related products or by-products are yet to be investigated.

2.3 Needs for Early Warning Systems

The need for an Early Warning System arises from the fact that the Qachqouch spring is a highly reactive spring that drains a mature karst catchment. Spring responses in terms of quality and quantity are highly pronounced (flow peaks and contaminant breakthrough as a response to precipitation events; Doummar and Aoun 2018, Allouch, 2020). The spring is used currently during baseflow, however, it can be used during high flow periods as alternative domestic water resources. Therefore, the absence of wastewater treatment plants and the presence of quarries, and limited agricultural practices pose a future risk for water consumption. It is worth noting that the implementation of an early warning system is in its very early stage and cannot be performed on this spring, given that the likelihood of contamination (bacteriological breakthrough) is relatively high in the absence of wastewater treatment plants. However, it could be applied to evaluate the potential arrival of high turbidity to inform the decision-makers in due time. In that case, the establishment of a strategy for the development of an EWS would be very useful for the Water Establishment and the treatment plant to ensure a better supply and management of the water resource.

2.4 Research to date to establish the components of EWS

The four key-components needed for the preliminary establishment of an EWS framework have and are being assessed on the Qachqouch spring: 1) flow and transport dynamics in the system, 2) sources of pollution and hazards, 3) monitoring system and data collection, and 4) legislative framework for water quality for comparison purposes.

1. The understanding of the flow dynamics of the water body was done through a flow numerical semi-distributed lumped model. Point source infiltration was assessed from tracer experiments and precipitation event analysis:
    a) Contaminants are channeled from various point source infiltration (autochthonous and allochthonous) leading to a variable response at the spring (the type and concentration of contaminants). The latter is also reflected in different turbidity peaks depending on the saturation of the aquifer. High turbidity is found to be associated with river infiltration (Doummar and Aoun, 2018b).
    b) A tracer experiment performed under different flow conditions reveals a connection with the El Kalb River.

2. The sources of pollution related to the land use/cover need further assessment based on the detected type of pollutants and evaluation of potential sources of pollution in the catchment.
    a) **Contaminants type:** The assessment of pollutants in the El Kalb River was performed during high flow and baseflow (in terms of concentration and loads). Additional assessment of other types of pollutants (heavy metal, industrial, petroleum products, pesticides, and fertilizers are still to be assessed, even if relatively very negligible in this rural set up)
    b) **Hazards and risk GIS map:** A mapping of the pollution sources was done preliminarily (Quarries, and hospitals). Domestic wastewater pits and gas stations were not mapped.
    c) **Relationship between measured parameters and contaminants:** An association between indicator parameters and contaminants was done partially and requires further investigation over the long run (correlation of Chloride with wastewater breakthrough, River input with turbidity, the arrival of newly infiltrated waters correlated with EC). An additional qualitative analysis of collected time series (2019-20) and particular events to detect potential indicators for different types of infiltrated waters.

3. Infrastructure for data collection and transmission to ensure continuous monitoring and collection of real-time data:
    a) Monitoring system and collected parameters relevant to EWS: A multi-parameter probe measuring turbidity (TU), Electrical Conductivity (EC), temperature (TEMP), and water level (WL) is installed in the spring.
    b) A telemetry unit is supposed to be shipped in January 2021 to secure transmission of real-time data via File transfer protocol (*ftp*).

c) The development of a dashboard for the display of processed data and real-time transfer of data (supported by alternative funds)

4. Assessment of guidelines for pollution identification (local and international guidelines for potable water)

2.5 Preliminary collected data (2019-2020)

Data collected at the spring relevant to the establishment of an EWS are as follows:

1) Data time series collected in-situ to date include in addition to *flow rates (Q), Electrical Conductivity (EC), Temperature (T), Turbidity (TU;* Figure 2.3*), Dissolved Oxygen (DO;* Figure 2.4*).*

2) Particle size analysis is measured in collected samples every 3 days with a Coulter Counter (aperture (100) for grain size varying between 2 and 60 µm; Figure 2.5, Figure 2.6).

3) Major ions are also analyzed in individual samples using Ion Chromatography (IC). Bacteriological analysis was performed Jan- March 2020 (Figure 2.7).

4) Stable isotopes are also analyzed in grab samples (collected every 3 days) using a PICARRO isotopic analyzer L2130-i cavity ring-down spectrometer (CRDS)

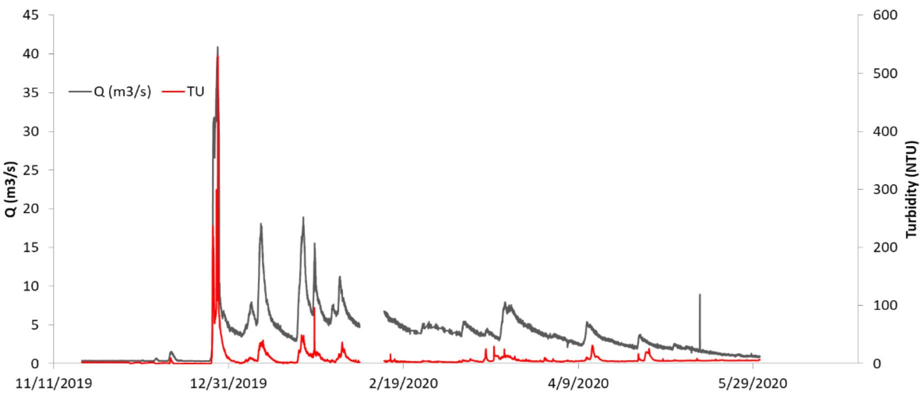

Figure 2.3. Comparison between flow rates and turbidity breakthrough observed at the spring

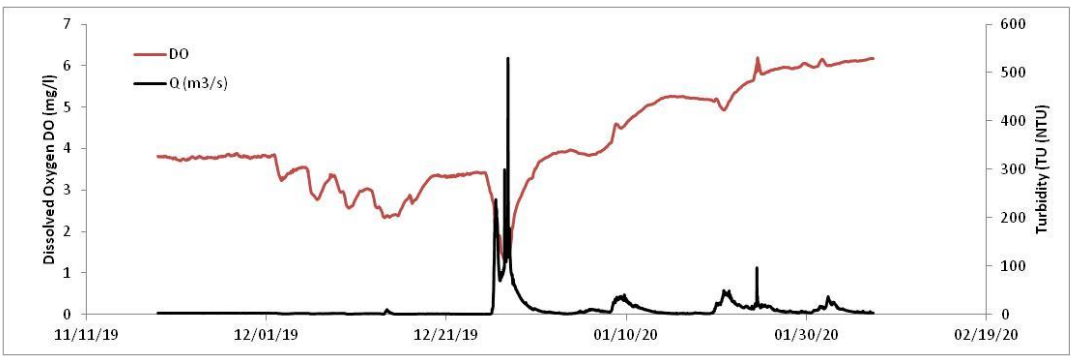

Figure 2.4 Comparison between turbidity breakthrough observed and dissolved oxygen measured at the spring

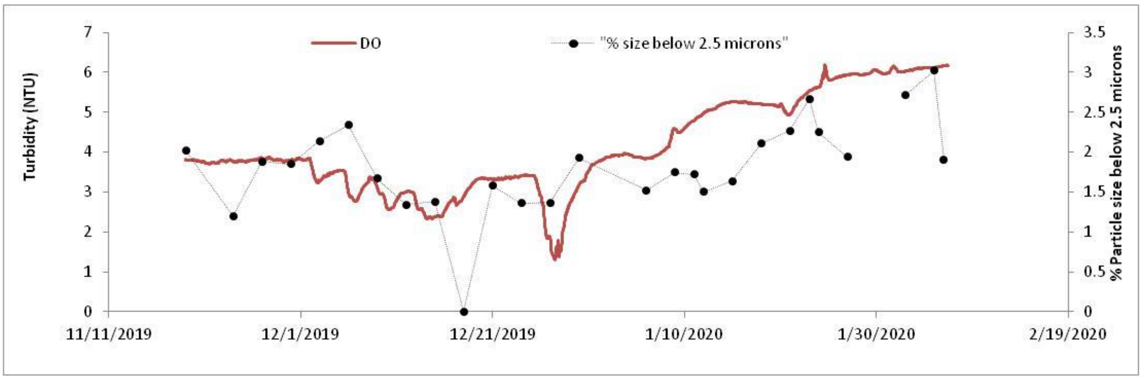

Figure 2.5. Percent of particles below 2.5 microns in comparison to Dissolved Oxygen (DO)

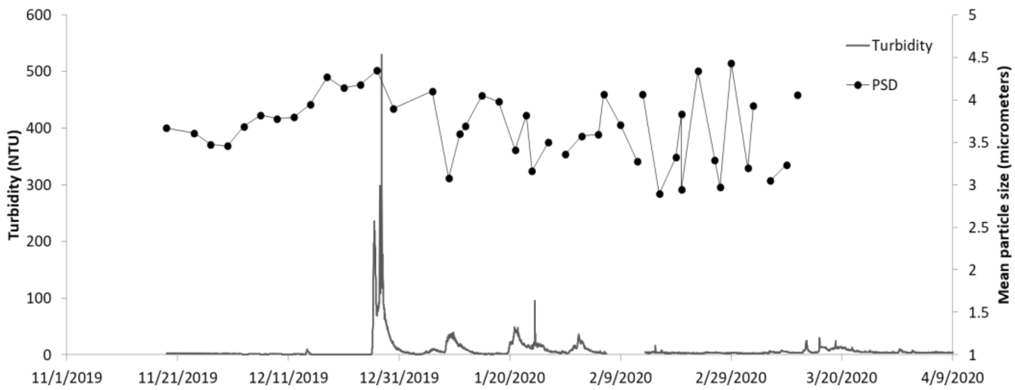

Figure 2.6. Mean particle size analysis in comparison to turbidity signals observed in spring responses

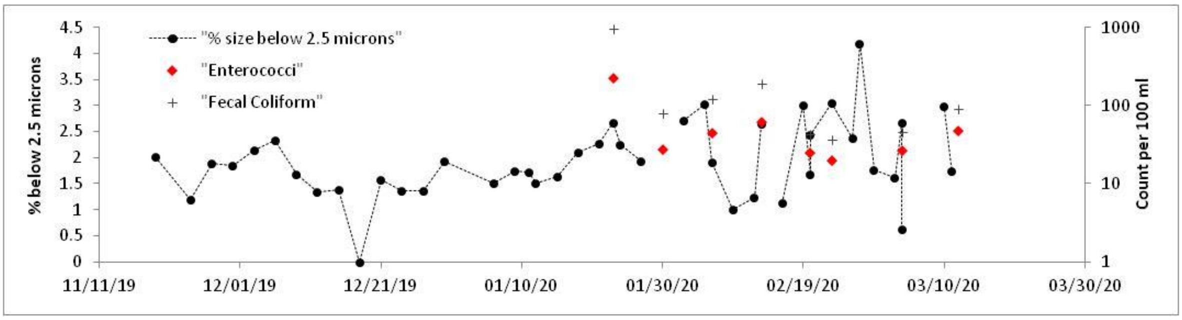

Figure 2.7 Correlation between bacteriological analysis and % of size below 2.5 % over a limited period of time (January-March 2020)

2.6 Data management

The flow and transport models to be developed in WP4, could be implemented as a decision tool to evaluate the risks for spring contamination with specific contaminants from different sources.

The proposed data management plan to support an EWS lies in the collection of data via a data logger, that will be transmitted via ftp (with the telemetry unit with gprs). The data will be processed and stored on the server and displayed on the dashboard. The processing of the data requires the evaluation of parameters that can be used as indicators in the absence of real-time monitoring of particle size, bacteria, etc.). This scheme is pending the installation of the telemetry unit planned end of January.

2.7 Limitations and data gaps

1) A lack of long-term measurements of chemical and bacteriological parameters for an appropriate assessment of spring responses. An additional interruption of data collection and analysis for several months in 2020 because of Covid-19 pandemic.

2) A lack of real-time measurements of bacteria, TOC, and particle size, or other relevant parameters or indicators.

3) The relatively poor characterization of the pollution sources at a catchment scale especially for private wastewater effluents.

4) The complexity of the flow dynamics and poor subsurface characterization of the Qachqouch spring system.

2.8 References

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3 Ubrique test site (case study Spain)

3.1 Introduction

A karstic area of 26 km^2 in the surroundings of Ubrique village (Cádiz province, S Spain; Fig. 3.1) has been selected for developing and testing an *Early Warning System* (EWS) due to its favourable hydrogeological features. These are: the village dependence of karst groundwater for drinking water supply, a well developed karst network that favours groundwater fast flows, and the most important one, the significant allogenic recharge which enters the aquifer through a swallow hole.

In rural karst areas, such as Ubrique test site, different sources of organic contamination (commonly linked to livestock activities) are observed. Previous investigations carried out in this area since 2012 (Sánchez et al., 2016; Martín-Rodriguez et al., 2016; Sánchez et al., 2017; Marín et al., 2020) allowed to identify different issues related to groundwater quality as well as exploitation of main drainage points intended for drinking water supply. Thus, part of its related hydrogeological infrastructure and monitoring network just needed to be updated and adapted to KARMA project execution.

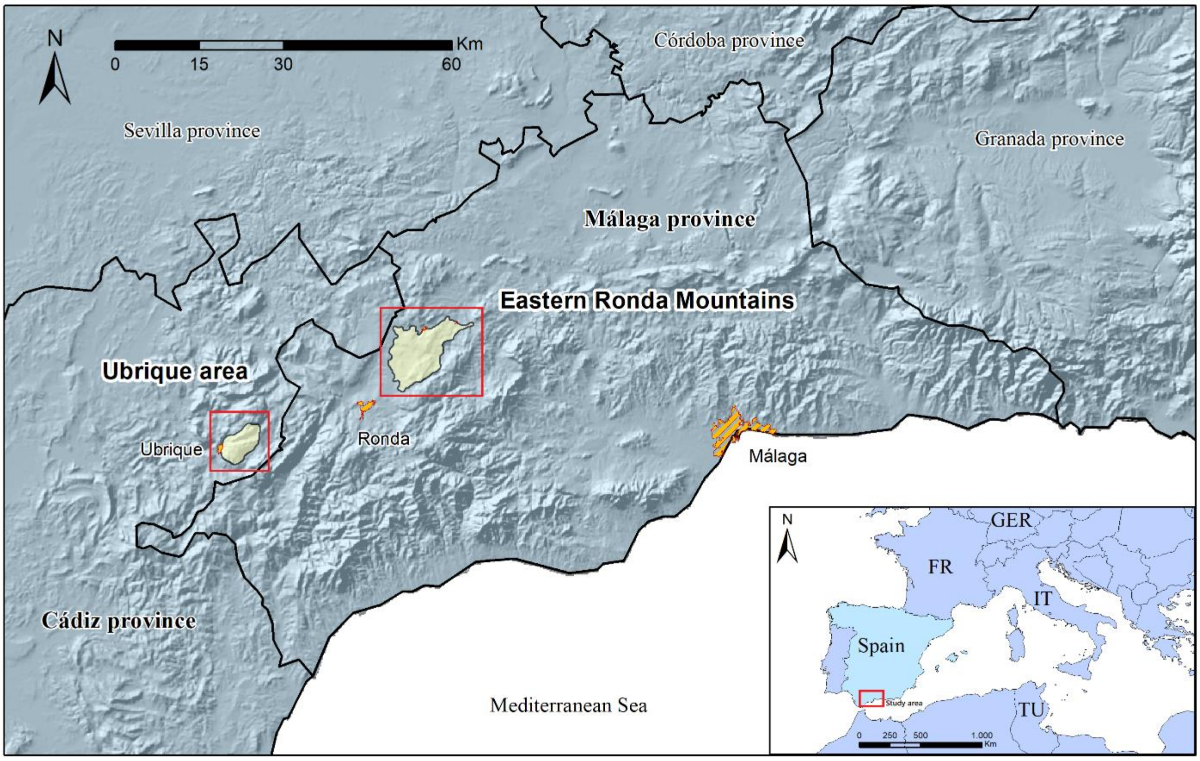

Figure 3.1: Geographical context of the Eastern Ronda Mountains and Ubrique area.

Ubrique area is composed of the homonymous mountain (or Sierra, Spanish term), which constitutes an entire aquifer system (Fig. 3.2), and a small shaft catchment basin. Mean annual precipitation in this area has been estimated around 1350 mm, however, it can variate depending on the altitude and sector from 900 mm to 1800 mm in the highest zones (Sánchez et al. 2016).

The relief in this area presents an alignment NE-SW with steep slopes that range from 800 to 1400 m.a.s.l. From a geological standpoint, Ubrique test site is characterized by the following geological formations (from the bottom to the top): Triassic evaporites with clays, Jurassic limestones (upper) and dolostones (lower) -500 meters thick-, and Cretaceous-Paleogene marly-limestones (Cruz-Sanjulián, 1974; Martín-Algarra, 1987). The geological structure is defined by anticline folds, in which core limestones and dolostones can be found, and synclines matching with depressions constituted by younger marly-limestones materials.

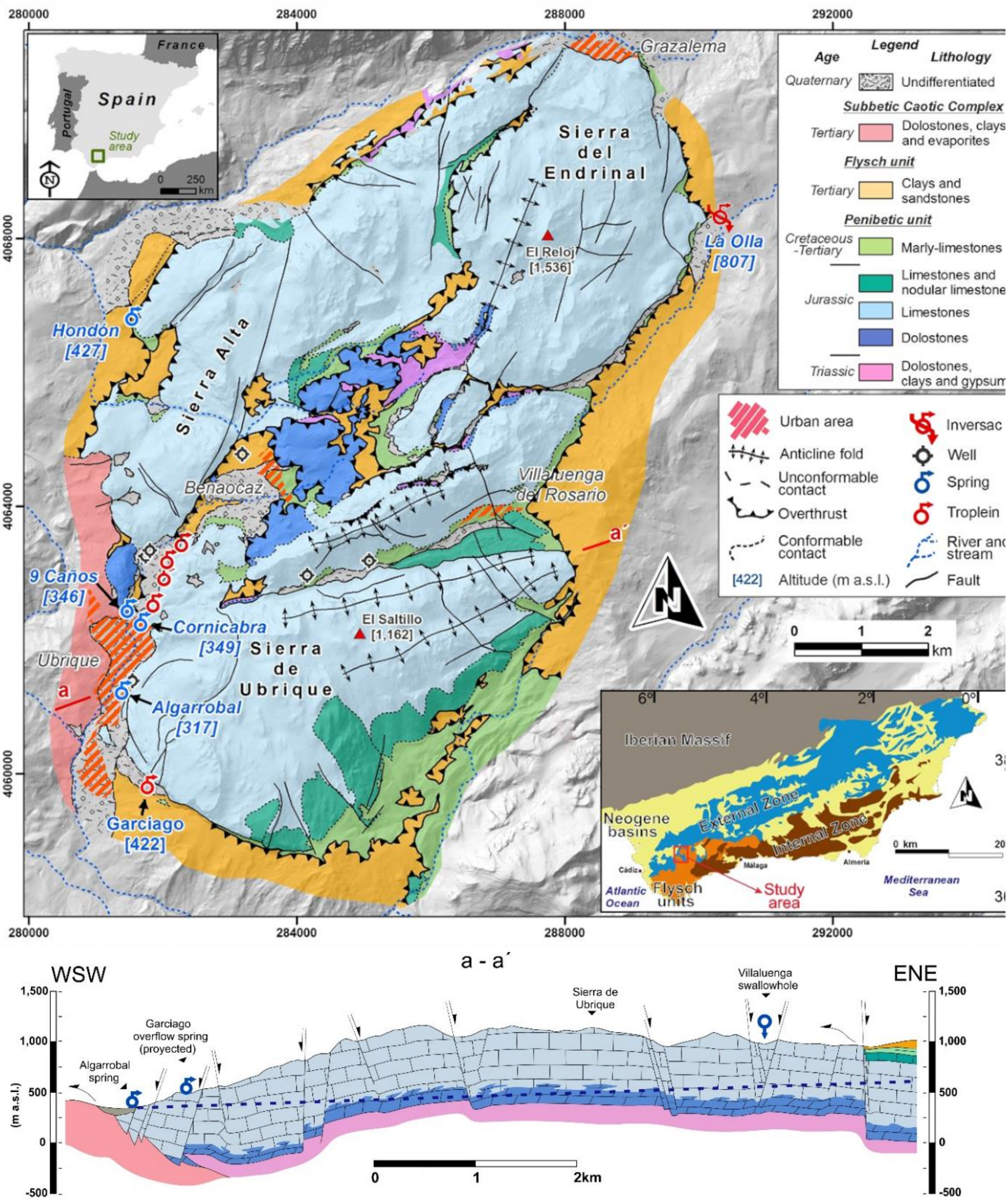

Figure 3.2: Location, geological map and SE-NW oriented hydrogeological cross-section of Ubrique aquifer (modified from Marín et al., 2020).

From a hydrogeological point of view, the study site constitutes a fractured and karstified aquifer system where discharge occurs through the springs of Cornicabra (349 m a.s.l.) (Fig. 3.3a) and Algarrobal (317 m a.s.l.) (Fig. 3.3b) (Martín-Rodriguez et al., 2016), located in the western border of the study area between the permeable carbonate rocks and the impervious layers (mainly Cretaceous-

Paleogene marly-limestones). Recharge in this area is produced in two ways, diffuse infiltration from rainfall through carbonate outcrops and concentrate flow from a small neighbour catchment which enters the system through the Villaluenga shaft. The use of artificial tracers allowed to verify hydrogeological connection between direct infiltration points and the two main springs.

3.2 Characteristics of the karst groundwater supply

The population of Ubrique nowadays is established around 16.500 people (even though it is usually increased in summer time) and the average daily water consumption intended for inhabitant has been estimated in 110-160 litres (based on tap water meter readings), this leads to a mean value of 2.300 m^3 consumed per day in the whole village. The two springs mentioned in the previous section constitute a fairly high percentage (80-90%) of drinking water intended for human consumption in Ubrique village, and in them, the main hydraulic infrastructures are arranged. Both catchment points are equipped with catchment houses, chlorination rooms and pipelines directly connected to the tanks and the village distribution network.

Figure 3.3: A) Inside of the Cornicabra spring catchment house with the autosampler and the field fluorometer; B) Algarrobal spring. Photos taken by: Jaime Fernández Ortega.

The water company also disposes of alternative groundwater catchment points located in a different and small hydrogeological system from the studied one called Sierra Alta aquifer, to solve temporary supply issues during the summer season under low flow conditions. These ancillary catchment points consist on two boreholes (Rano I and Rano II) and one karst spring called Nueve Caños, which are located in the northern part of the village. The complete urban water supply network (Fig. 3.4) and its related hydraulic infrastructure is then constituted by 5 groundwater catchment points (2 springs and 3 boreholes), 3 water tanks and 1 pumping station. Three different distribution areas are established depending on the dwellings' elevation with respect to tanks, so that drinking water is finally distributed by gravity.

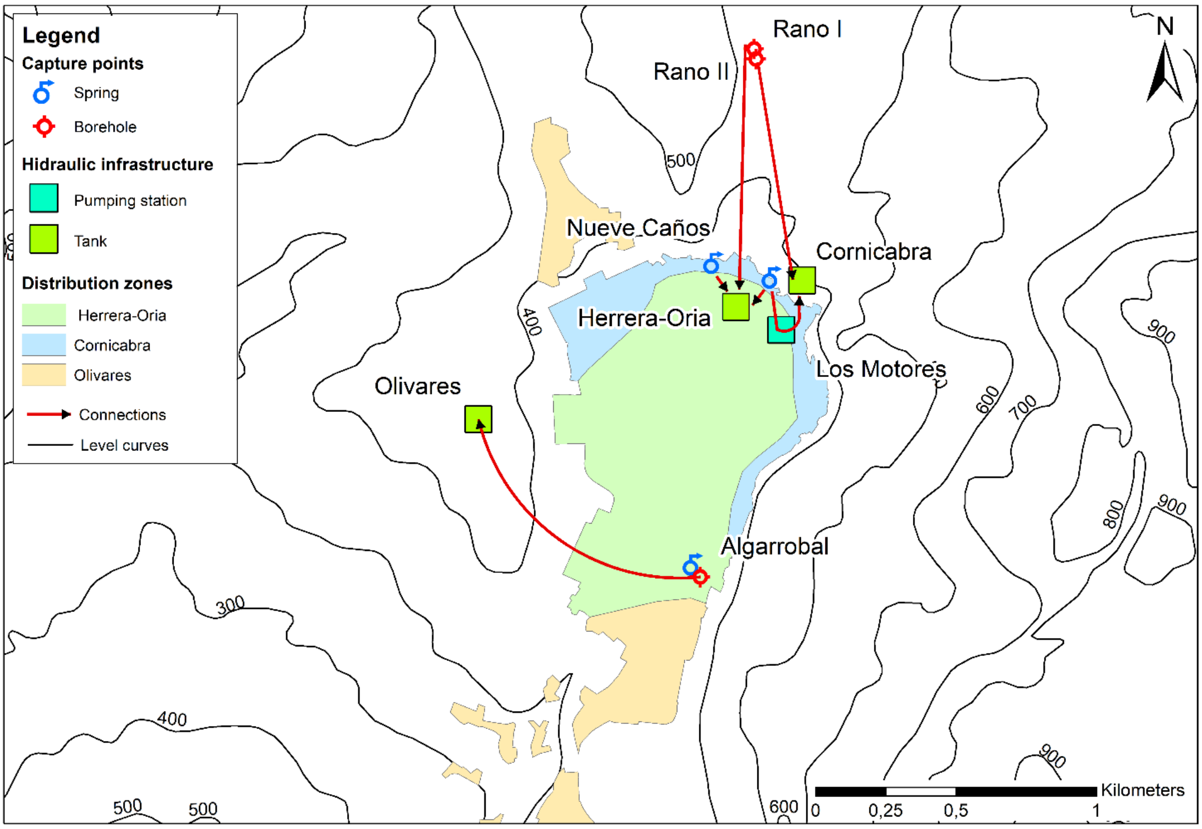

Figure 3.4: Scheme of the urban water supply network.

3.3 Groundwater capture points and water treatment system

In Spain, Royal Decree 140/2003 establishes on 1 mg/l the maximum amount of residual chlorine present in drinking water, however the defined threshold to qualify water as unfit for human consumption is 5 mg/l (which indicates a wrong treatment procedure). In the case of Ubrique water treatment system, residual chlorine concentration is kept between 0,2-0,8 mg/l, in that way, groundwater is treated with drinkable sodium hypochlorite 14% (CAS: 7681-52-9) and the manufacturer recommends to add it in the following concentrations (Tab. 3.1):

INITIAL BLEACH CONCENTRATION	WATER QUANTITY TO TREAT		
Free chlorine per litre	10 litres	100 litres	1000 litres
150 g/l	4 drops	2 ml	20 ml
100 g/l	6 drops	3 ml	30 ml

Table 3.1: Recommended free chlorine volumes for water treatment.

The **capture points** are distributed as follows:

- Cornicabra spring: groundwater outflow is protected by a small catchment house (Fig. 3.5a) where water is treated and directly driven by gravity to the water supply network. A small volume of water is derived to the Cornicabra tank through the pumping station. Sodium hypochlorite is added from the chlorination through a small conduit and poured into the water at the entrance of the distribution pipeline. A filter grid is also set at the same place to prevent the entrance of solid particles or fatty substances.
- Algarrobal spring: groundwater is pumped at night (usually from 2 am to 7 am, depending on drinking water demand) through a borehole located just 20 meters upwards from the spring. At this catchment house (Fig 3.5b), groundwater is also treated and then transported through a pipeline to Los Olivares tank.
- Nueve caños spring (auxiliary): the water is driven by gravity to the Herrera-Oria tank and sometimes used to increase distribution system pressure when Cornicabra spring discharge is not enough.
- Rano I (auxiliary): the water is as well treated at the catchment point and then pumped to the Herrera-Oria tank. This borehole is activated only when none of the springs outflow is enough.
- Rano II (auxiliary): it is located quite close to the previous borehole but then pumped to the Cornicabra tank. This borehole is automatically activated when Cornicabra tank water level decreases under a certain threshold.

Water tanks and pumping station:

- Cornicabra: this water tank is located just 50 meters above the same-called spring and can store up to 520 m^3. It mainly receives water from Cornicabra spring and the Rano II borehole and Nueve Caños spring when necessary. It distributes drinking water to the upper-northern part of the village.
- Herrera-Oria: this water tank can store up to 500 m^3 and it is located just 60 meters downstream from Cornicabra spring. When used, it receives water from the last mentioned and Nueve Caños spring in order to increase pressure (+0,5 bar) of the distribution system. The Rano II borehole is also connected to this tank, which provides drinking water to the lower part of Ubrique.
- Los Olivares: this round shape water tank can store up to 3.000 m^3 and it is located close to the southern access to Ubrique village. It only receives water from Algarrobal spring and it is used to supply the upper-eastern parts of the village.
- Los Motores pumping station: it collects the water from Cornicabra spring and Herrera-Oria tank (when necessary) and pumps it upwards to Cornicabra tank.

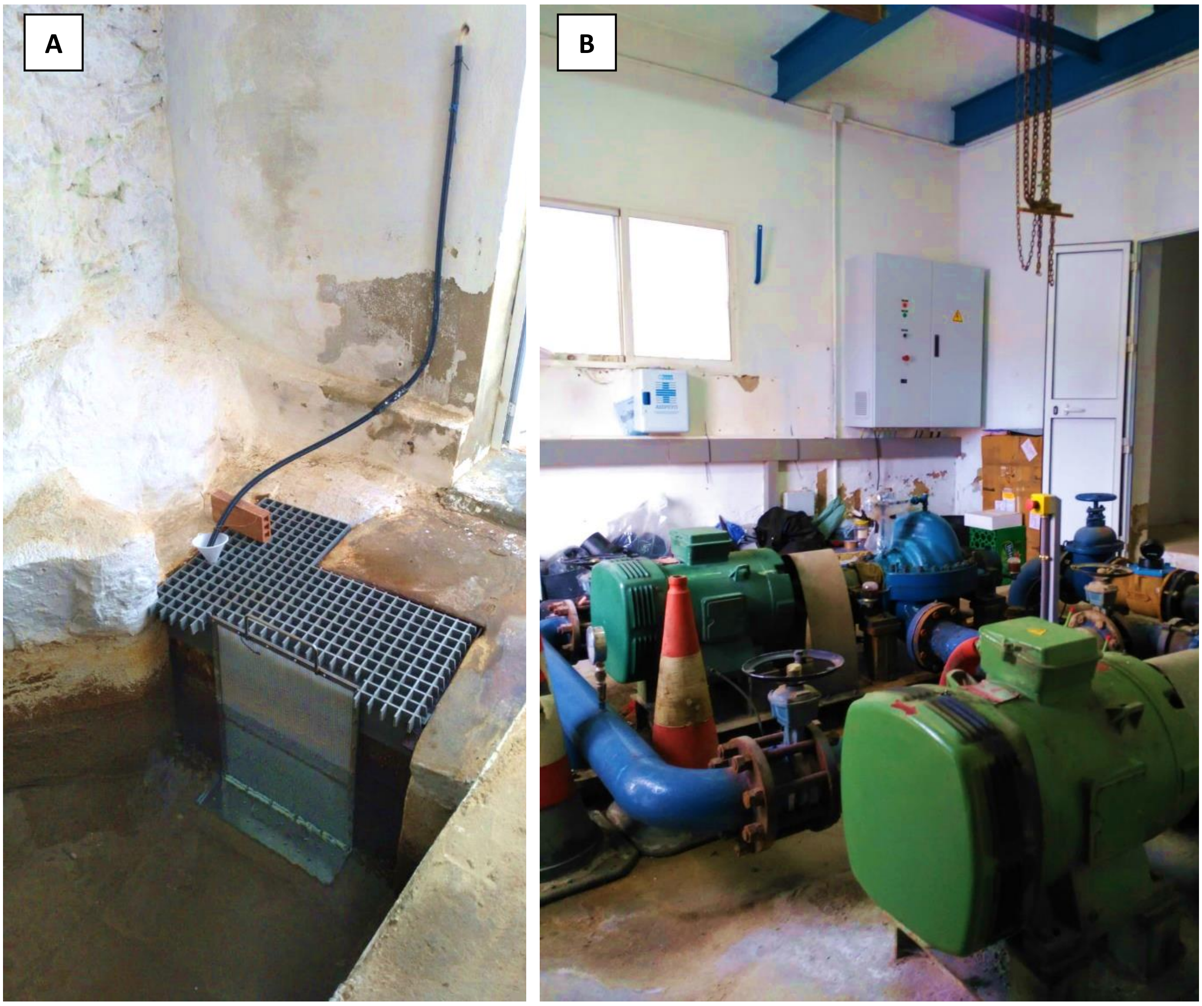

Figure 3.5: A) Water derivation to the tank and treatment at Cornicabra spring. Photo taken by: Jaime Fernández Ortega. B) Inside of the catchment house and pumping system at Algarrobal spring. Photo taken by: José Francisco Martín Rodríguez.

3.4 Water quality issues to justify the implementation of an EWS

As mentioned in the first section, Ubrique area is the most suitable test site for the development and implementation of the EWS due to its specific features that promote short-term contamination events at the two main springs of the aquifer. This temporary low water quality conditions hinder the total exploitation of groundwater intended for human consumption.

Sierra de Ubrique aquifer presents a pretty high development of karst network, different shafts and galleries have been widely explored and documented by speleologist since the last 50 years (Romero, 1970; Pedroche et al., 1980, Mendoza, 1992). Specifically, Villaluenga Shaft (Fig. 3.6) presents a depth of 130 meters until the first siphon and 200 meters until the second one (of a total of 5 siphons explored). This feature in addition to concentrated flow that enters through the shaft, favours fast groundwater flow (>100 m/h) to the springs located in the eastern part of the aquifer.

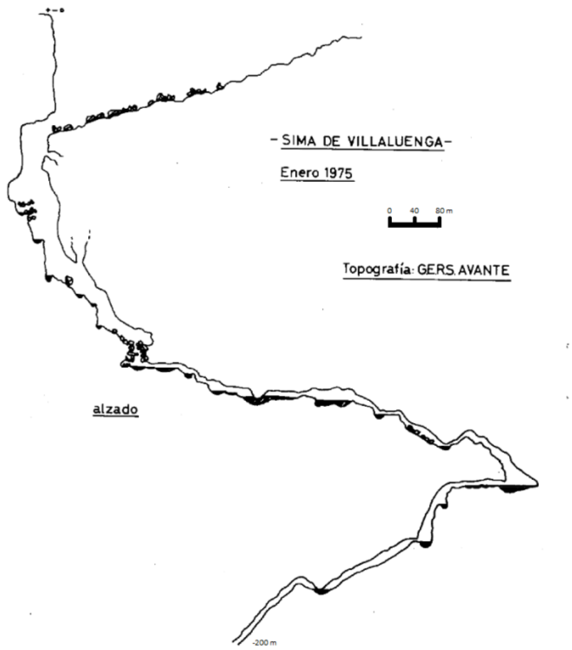

Figure 3.6: Cross section of Villaluenga Shaft (taken from Pedroche et al., 1980).

The catchment area feeding Villaluenga shaft (Fig. 3.7) displays a total surface of 3 km^2, however, only 35% corresponds to impervious layers. Despite the small draining surface, when stormy rainfall occurs, it can generate enough runoff to wash the soil from the surrounding areas and carry the fecal remains of the livestock (Fig. 3.8a). This common situation leads to the activation of the Albarrán stream, which transports the contaminant load in addition to the effluent from the waste water treatment plant (which is directly poured into the rivedbed) (Fig. 3.8b) and inorganic sediment particles (specially increasing the contaminant load after a long dry period, e.g. summer season).

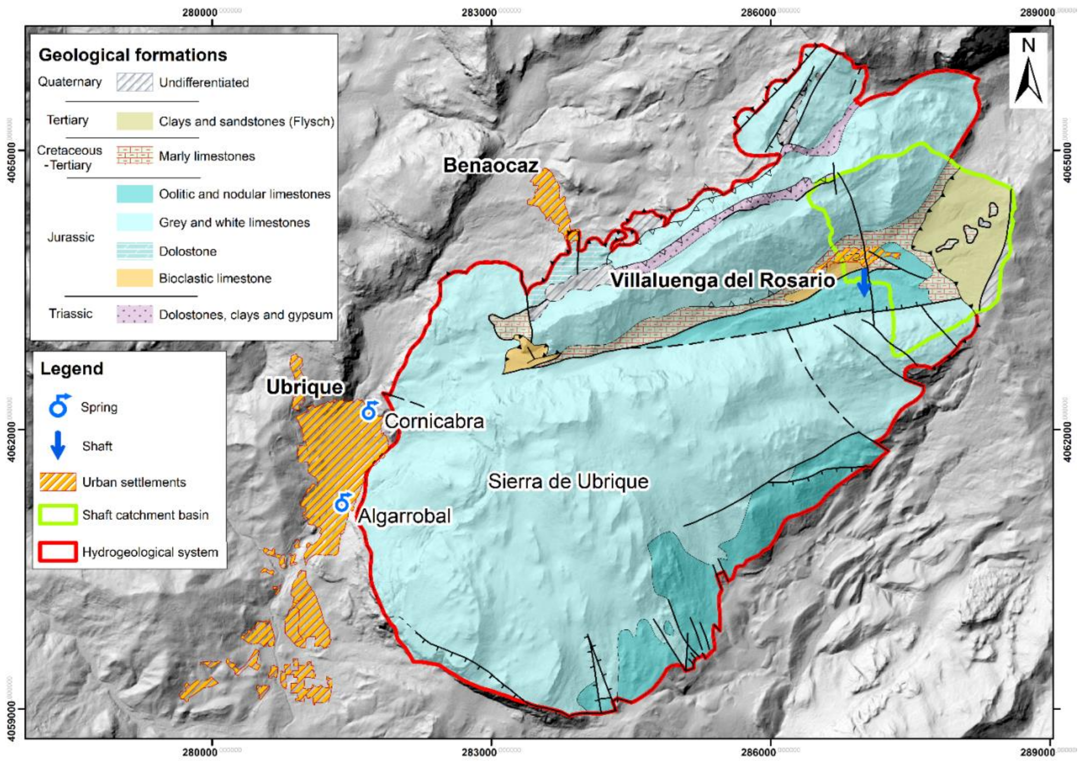

Figure 3.7: Limits of the hydrogeological systems and shaft catchment basin.

The wastewater treatment plant of Villaluenga del Rosario village (Fig. 3.9) is located just 150 meters far from the shaft and it is well known that its effluent does not accomplish with the minimum quality requirements established by Directive 91/271/CEE. As an example, the effluent sample taken on 16/11/20 showed 319,2 mg/l of Total Nitrogen and the maximum is set at 15 mg/l.

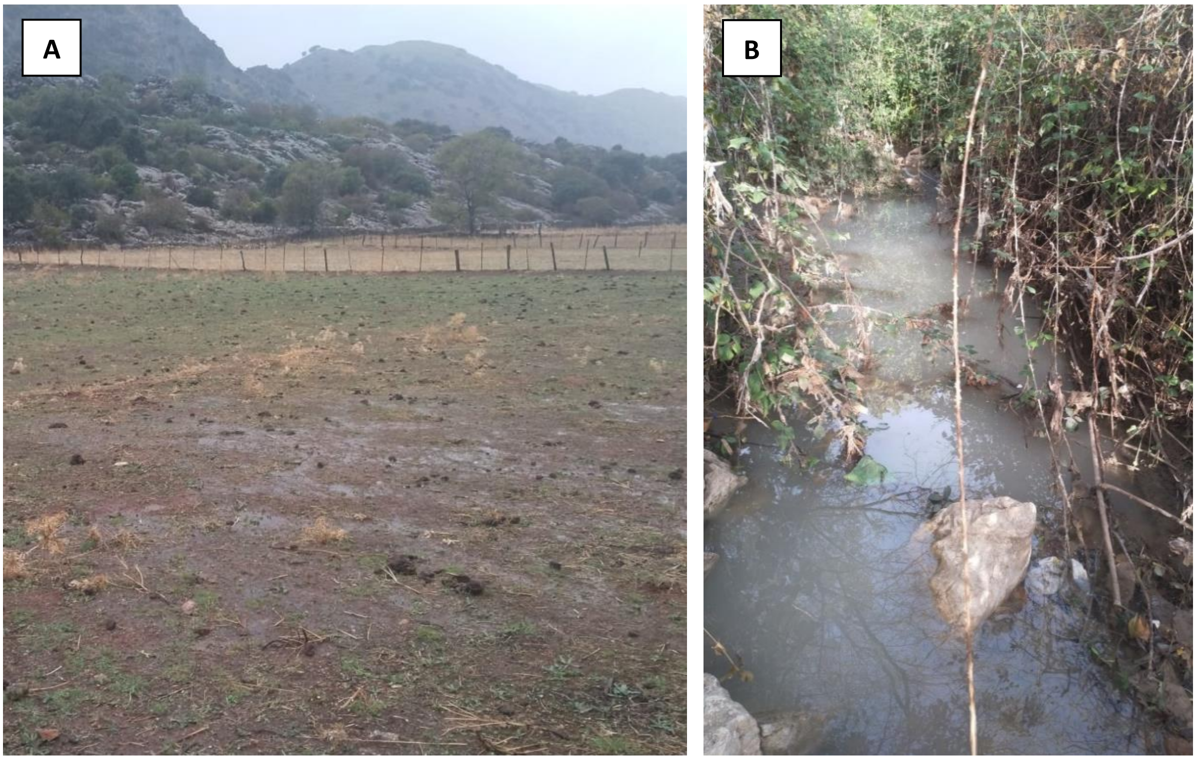

Figure 3.8. A) Remains of livestock activity on a rainy day, B) Wastewater treatment plant effluent. Photos taken by: Jaime Fernández Ortega.

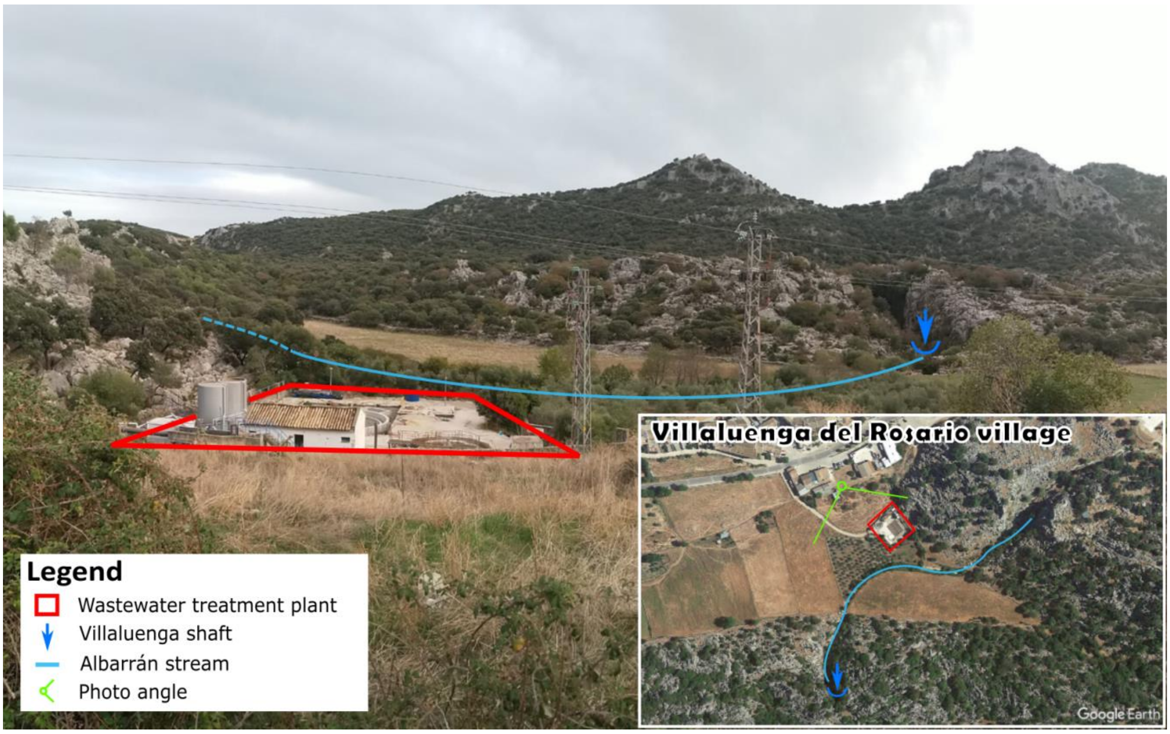

Figure 3.9: Panoramic view of the Villaluenga shaft and the waste water treatment plant. Photo taken by: Jaime Fernández Ortega. Reference photo taken from Google Earth.

One of the most important steps for the development of the EWS is the contaminant characterization. In this case, both organic and inorganic contaminants can be identified. The fact that the wastewater treatment plant receives the whey of six cheese factories located in the village of Villaluenga del Rosario elucidates one of the main contaminant sources. This cheese is made of goat and sheep milk. Ovine and caprine milk proteins are present on the cheese whey, and they contain different amino acids, among which, tryptophan is present in all of them (Amigo and Fontecha, 2011; Ramos and Juarez, 2011). Fecal remains and the application of manure in the surrounding fields also act as a source of nitrate (NO_3^-) and *E. Coli* (Drew and Hötzl 1999; Boyer and Pasquarell 1999). Further genetic and contaminants of emerging concern analysis will be performed to clarify and elucidate all the substances and microorganisms present in the inflow and outflow of the system.

This special condition, as described in the first paragraph, results in high turbidity and associated bacterial contamination discharge events (Fig. 3.10) which endanger Ubrique village drinking water supply, hindering the total exploitation of the available resources since in these situations it is not possible to pump water from the aquifer. In Spain, Royal Decree RD140/2003, February the 7th, establishes the sanitary criteria for the quality of water for human consumption among which are the parameters that generate this kind of problems in Ubrique drinking water.

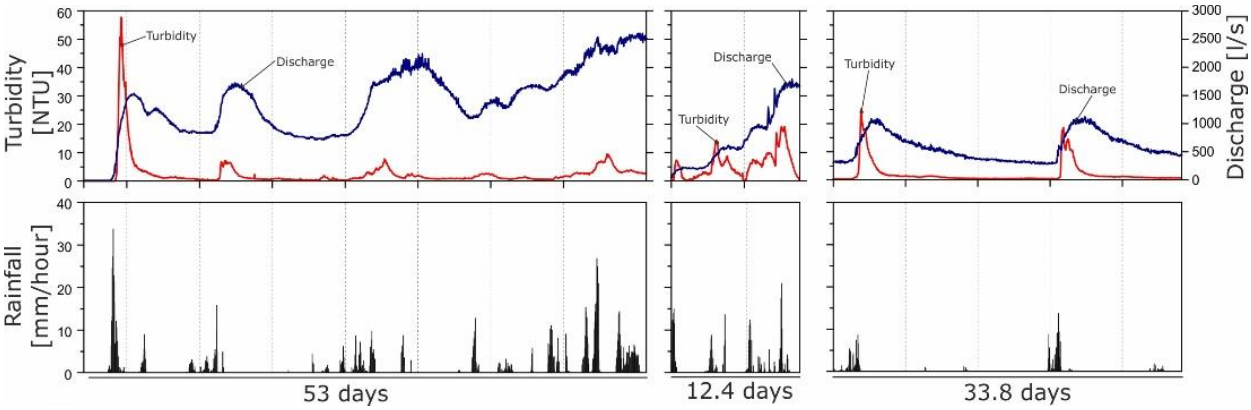

Figure 3.10: Time series of discharge and turbidity in Algarrobal spring after several rainfall episodes during an approximately 2 month-time window. (Taken from: Marín et al., 2020)

Figure 3.11 shows both springs intended for drinking water supply under high turbidity conditions after a storm event in March 2018. Turbidity values can reach 60-80 NTU and hinder the total exploitation during days after the recharge event.

The presence and transport of pathogenic microorganisms (protozoans, viruses, bacteria) in fissured and karst aquifers has been widely studied in the last decades (Champ and Schroeter, 1988; Rose et al. 1991; Personne et al. 1998; Mahler et al., 2000; Pronk et al. 2007). Previous studies have demonstrated the survival potential of microorganisms when they are associated to particulates (Palmateer et al., 1993; Pommepuy et al., 1992), acting as a vector for transport of other contaminants because of the ability of bacteria to sorb onto particulates (Mahler and Lynch, 1999; Mahler et al., 1999). Also, previous researches verified the relationship between tryptophan-like fluorescence and labile organic carbon and microbial activity (Cammack et al., 2004; Hudson et al., 2007; Lapworth et al., 2007; Hudson et al., 2008, Sorensen et al., 2015). Thus, the conjoint utilization of L-tryptophan, turbidity and particle distribution emerge as a promising indicator parameter of bacterial activity and contamination events.

A

B

Figure 3.11. A) Algarrobal spring during high turbidity conditions. B) Cornicabra spring during high turbidity conditions. (Photos taken by: José Francisco Martín Rodríguez in March 2018).

3.5 Typology and monitoring set-up of the EWS

An EWS consist of an integrated system for monitoring, analyzing, interpreting, and communicating monitoring data, which can then be used to make decisions that are protective of public health and prevent inconveniences to the users (U.S. EPA, 2005). Grayman et al. (2001) presented a wide review of surface water models for use in EWSs. In September 2017, the United States Environmental Protection Agency (EPA) published a State-of-the-Science report on Drinking Water Treatment Source Water Early Warning System in which the origins and first developments of surface water EWS are described (U.S. EPA, 2017). This review presents a taxonomical classification of EWS models based on complexity: physically-based, GIS-based, data-driven models and simplified modeling techniques. Three basic components of a EWS model commonly include: a flow module, a water quality transport module and a fate module. The flow module describes the movement of water; the water quality transport module describes the processes by which the contaminant concentration moves and changes due to the hydrodynamic forces; and the fate module describes the impacts of physical, chemical and biological processes on the form and concentration of the contaminant.

Some of these techniques and applications can be adapted to groundwater and karst hydrogeology since it shows some similarities with surface hydrology. In that way, the EWS proposed for Ubrique test site could be then considered a junction between data-driven and physically based models, as it will integrate: (1) modeling tools that use mathematical equations which represent real-world physical-chemical processes (e.g. karst groundwater flow and spring discharge; sediment load, pathogens or solute transport) and (2) a database of spring discharge, physical-chemical and biological water parameters as well as meteorological information (Tab. 3.2) which constitute an essential part for the application of Big Data analysis techniques and Artificial Neural Networks (ANN) (Zhang et al. 2018).

In order to acquire such database at Ubrique test site, different approaches are being applied such as laboratory analysis of groundwater samples (as described in Table 3 from Deliverable 3.1.), the continuous record of natural responses and the execution of dye tracer test at different hydrodynamic conditions. Three key karst features are then being fully monitored: Cornicabra and Algarrobal springs and Villaluenga shaft, in addition to the weather station located 1021 m.a.s.l. in Sierra de Ubrique.

<table>
<tr><th>Parameter</th><th>Time resolution</th><th>Device</th><th>Location</th></tr>
<tr><td rowspan="2">EC and temperature</td><td rowspan="4">15 min</td><td>HOBO U24 logger</td><td rowspan="10">Algarrobal spring

Cornicabra spring

Villaluenga shaft</td></tr>
<tr><td>GGUN</td></tr>
<tr><td>Inflow/outflow</td><td>Odyssey logger for water level recording</td></tr>
<tr><td>Tryptophan*, turbidity, aminoGacid*, DOM</td><td>Field fluorometer GGUN - Albillia</td></tr>
<tr><td>Physical-chemical parameters (pH, Eh, Dissolved Oxygen and turbidity)</td><td rowspan="6">2-4 h (high flow)
6-12 h (intermediate flow)
1-7 days (low flow)</td><td>HACH multiprobe HQ40D and turbidity pocket meter 2100Qis</td></tr>
<tr><td>Groundwater / stream water samples</td><td>Autosampler- ISCO 3700</td></tr>
<tr><td>Soil/water Rn^{222} activity*</td><td>SAPHIMO – AlphaGuard PQ2000 PRO 4800</td></tr>
<tr><td>Particle size distribution</td><td>Klotz - Abakus mobil fluid touch</td></tr>
<tr><td>E. Coli concentration</td><td>IDEXX Colisure</td></tr>
<tr><td></td><td></td></tr>
<tr><td>Rain, air temperature and RH</td><td>15 min</td><td>Davis weather station</td><td>Sierra de Ubrique</td></tr>
</table>

Table 3.2. Parameter database source and monitoring set-up location (only at springs).*

The set of devices that compose the monitoring network has been selected to accomplish an array of desirable characteristics that make the EWS performance efficient:

- Sensitivity: according to the nature and range of the contaminant substances, the devices show a high accuracy to easily detect water quality variations through short time intervals.
- Affordable cost: a cost-effective balance has been performed in order to establish a relationship between the quality and accuracy of the data and the economical investment to obtain the data.
- Automation and remote data transfer: real-time information will be available for water company managers and decision makers to give a fast response to contamination events. A reliable and actualized telemetry system is essential for developing a robust EWS.

This process might be accomplished by a modular design that could be upgraded stepwise on a planned schedule to incorporate new technologies, as they become available (U.S. EPA, 2005).

3.6 Data management and statistical/numerical modeling

The groundwater EWS requires data management and visualization tools to support analytics and communication. The process of data analysis and the integration of the different model information in addition to weather forecast is complex. In order to achieve this task, different data treatment approaches are being adopted. The procedure for the implementation of the EWS in Ubrique test site has been recently described (Marin et al., 2021), and it will consist of 3 phases for its full execution (Fig. 3.12):

1. As stated in the previous sections, the continuous record of natural responses of the springs and the shaft, including both easy-to-measure and novel parameters, it is being carried out and will serve as a basis for numerical simulations.

2. Analysis of short/long-term data series through different statistical techniques in order to optimize the performance of the EWS: I) simple and multivariate statistical analysis, II) rain-runoff hydrodynamic modelling, III) allochthonous recharge (via Villaluenga shaft) vs. autochthonous (permeable outcrops) modelling in spring discharge and detailed hydrograph analysis, IV) modeling of dye breakthrough curves (BTC) under highly variable flow conditions, V) mass balance of harmful to health compounds, in order to stablish a hydrogeotoxicity characterization of different kinds of storm events, and finally VI) the application of properly adapted ANN-based and artificial intelligence algorithms to identify contamination events.

3. System launching with operational perspective and the assessment of selected operational Key Performance Indicators (KPIs). These KPIs comprises statistical metrics used to synthesize information on the efficiency and productivity of the measurement strategies carried out to optimize its operation. The optimization process will be continuous, as the system gets a feedback from the analysis of data acquired on each contamination event. The use of these indicators will allow to minimize False-positives/False-negatives alerts of the system as well as test the robustness and verify the continuous functioning of the EWS.

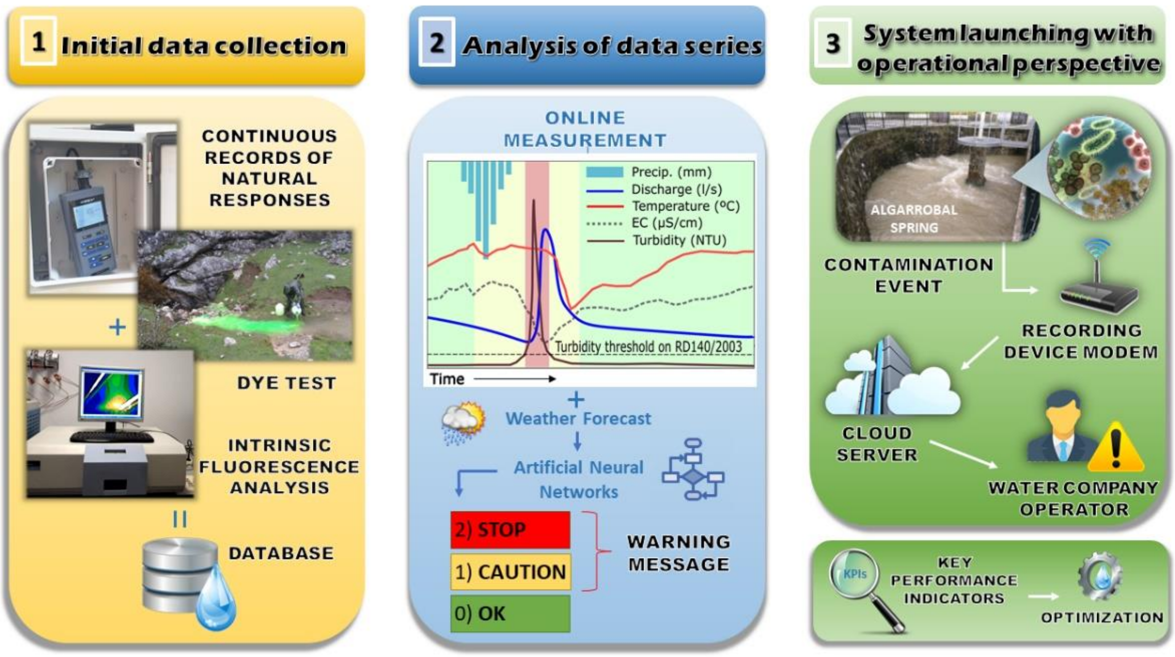

Figure 3.12. Implementation steps proposed for EWS at Ubrique test site. Taken from Marin et al., 2020.

3.7 First results: time series of selected chemical parameters recorded

Time series of both springs start on February 2020, however, the first recorded rain event dates from the third week of March. In the case of Cornicabra spring (Fig. 3.13), these four rain events produced a quite high increase of spring's flow discharge, where three high water intervals can be identified.

Despite the first recharge input did not generate a big spring response, regarding flow discharge, it caused a decrease of electrical conductivity, and on the contrary, an increase of tryptophan concentration. Just before the discharge peak, an increase of electrical conductivity and decrease of temperature can be observed, and after that, dilution process can be distinguished as the electrical conductivity diminishes due to the arrival of rain water, which also causes an increase of temperature. In correlation to the three discharge peaks, also three tryptophan peaks can be found, this means that despite the dilution processes, concentration of this substance fairly increases during high water conditions. These three tryptophan peaks display a "bitten" shape where two similar peaks can be differentiated, this could reflect the arrival of tryptophan remobilized from inside the system, and then, the tryptophan input through concentrated recharge through the shaft, which also coincides with maximum discharge. This may also explain why the time distance between the two peaks decreases through successive recharge events as they are produced during high-medium flow conditions.

After a really dry summer season, with no significant recharge events, higher mineralization of groundwater is observed and increased event more after a high single recharge event in first October which corresponds to piston effect, followed by a longer recharge event that causes a progressive dilution. These first recharge events of the hydrological year didn't produce any significant response at spring discharge, as field capacity in the recharge area was near the wilting point. However, Total Organic Carbon (TOC) concentration increases, indicating the arrival of diffuse infiltration from the soil, which is also related to the increase of ^{222}Rn gas concentration from the epikarst clays. During the rain episodes of November, a slight increase of discharge flow is observed, which is also related to a clear and continuous increase of turbidity and tryptophan. The effects of the recharge events are also clear through the observation of TOC, Total Nitrates (TN), sulphate and nitrate records, which could indicate a more significant arrival of concentrated recharge from the shaft, as ^{222}Rn is not as high as the previous event.

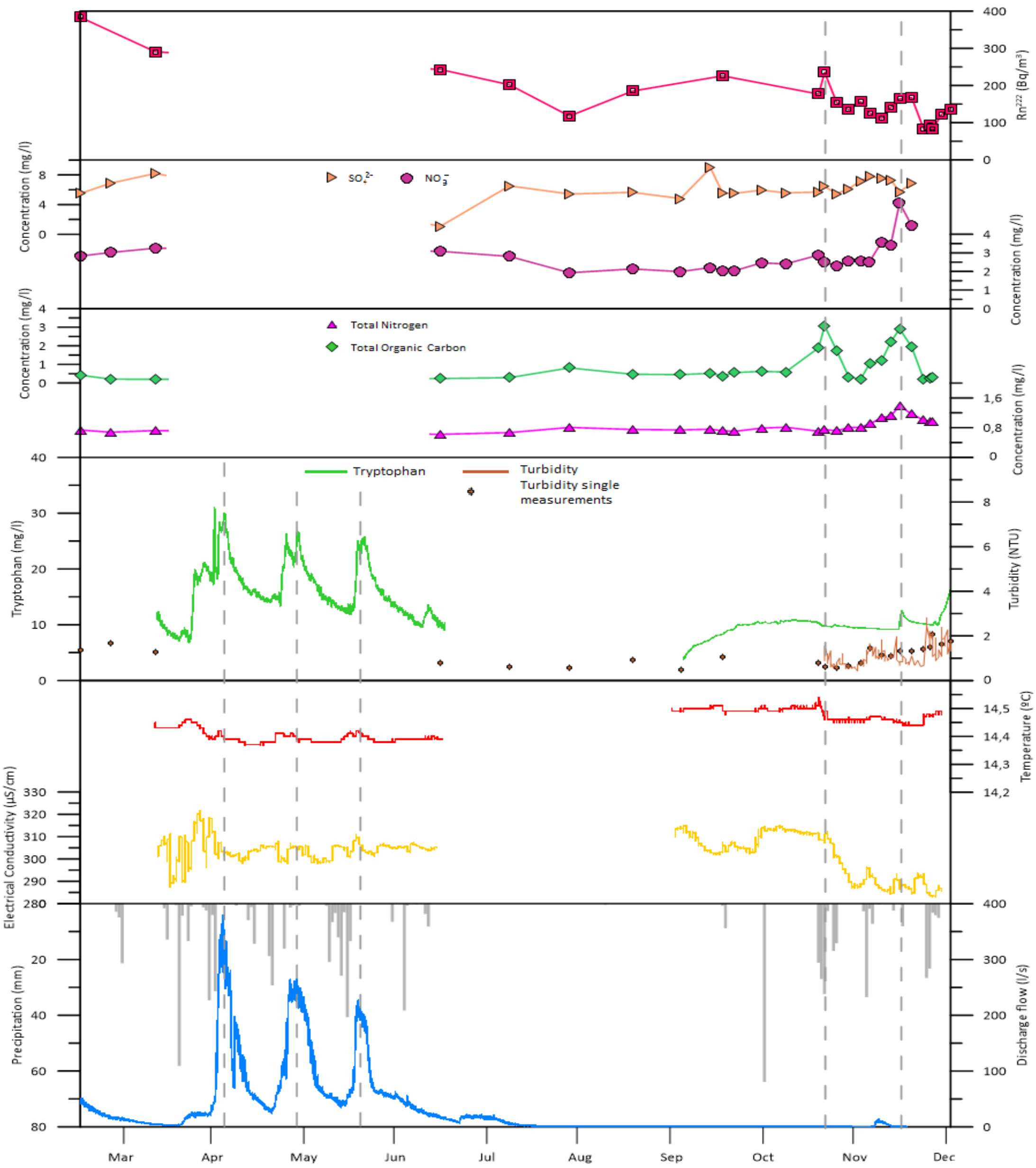

Figure 3.13. Time series of hydrodynamic and hydrochemical evolutions at Cornicabra spring.

Time series at Algarrobal spring (Fig. 3.14) are quite heterogeneous due to the effects of pumping. Algarrobal spring does not present such identifiable recharge events during spring time rains, but a continuous increase of discharge flow is observed. During the summer season, a continuous trend of increasing temperature, tryptophan concentration and mineralization is detected. The effect of the rain episodes in autumn is clearly identifiable at spring discharge (despite no big changes are recognized), temperature and electrical conductivity records. A decrease of electrical conductivity is related to an increase of groundwater temperature, indicating the arrival of newly infiltrated rainwater. An increasing concentration of ^{222}Rn gas is also observed, which is related to epikarst and soil infiltration. Chemical time series also show an increasing content of sulphate, nitrate and Total

Nitrate. In contrast to Cornicabra spring records, only one TOC peak at the end of the time series is detected.

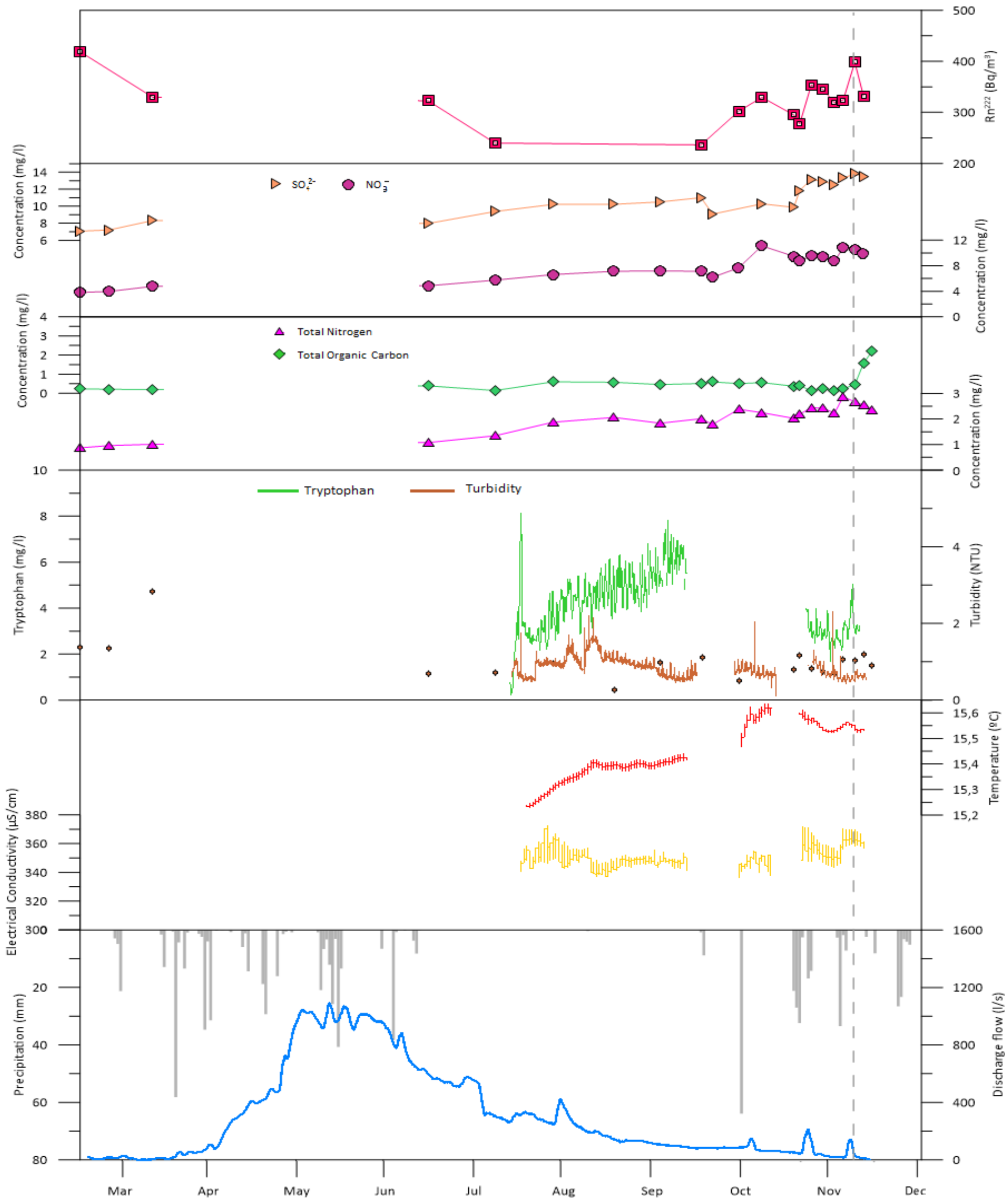

Figure 3.14. Time series of hydrodynamic and hydrochemical evolutions at Algarrobal spring.

3.8 Expected operational benefits from the implementation of an EWS

Regarding technical issues, exploitation process efficiency will be increased through the continuous monitoring of contaminant indicator parameters, which will allow to obtain a more detailed distribution of water quality through the time and to anticipate low water quality periods. Nowadays, contamination issues are only detected through visual inspection of groundwater turbidity, which could be not precise enough. In fact, the possibility that some contamination events do not exactly correlate with turbidity exist, and thus, volumes of groundwater unfit for human consumption could enter the system.

In the final operational phase, the information provided by the easy-to-measure recording probes for real-time monitoring and meteorological information will be transferred through telemetry systems to an online server in addition to meteorological predictions. Based on this web database, the algorithm continuously calculates the potential risk of a contamination event to occur and automatically validates its forecasting in near real time (15 minutes resolution) to warn about the risk of contamination for spring water quality (Grimmeisen et al. 2018, Zhang et al. 2019). The EWS will then display different warning messages to the water company operators and decision makers when the performance algorithm reaches a critical threshold, and thus, they could be assisted by the system in terms of optimizing operation works.

The implementation of this new online server presents multiple advantages: it will allow to have real-time knowledge about groundwater quality status and specific predictions for this test site, increasing the level of awareness when necessary. It is also intended to display system performance information with different access levels depending on the professional profile (drinking water users, water company technicians, decision makers…), therefore, the full implementation of the EWS in Ubrique village, depending on the new management strategy, will fairly improve the chemical water quality for urban supply.

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4 The Lez Karst Catchment (case study France)

4.1 Characteristics of the karst groundwater supply

The Lez spring (65 m.a.s.l), is located about 15 km north of Montpellier. It is of Vauclusian-type with a maximum discharge of approximately 15 m^3/s. However, the discharge at the Lez spring is generally small or nil due to the active pumping management of the aquifer, which supplies about 30 to 35 Mm^3 of water per year to the metropolitan area of Montpellier (340 000 persons). After a period during which only the natural overflow of the spring was used (from 1864 to 1965), water was pumped in the spring down to -6.50 m below the overflow level of the spring (65 m.a.s.l). On June 1981, a DUP authorized the construction of an underground pumping plant which was operational on December 1982 (Fig. 4.1). Four deep wells were drilled and reached the karst conduit feeding the spring, 48 m below the overflow level of the spring (17 m.a.s.l). Pumping these wells allows up to 1700 l/s to be withdrawn under low-flow conditions (with an authorized maximum drawdown of 30 m), while the average annual pumping flow rate is about 1100 l/s (1988–2009). This type of management is possible as long as the mean pumped flow rate does not exceed the mean annual discharge of the spring that is about 2200 l/s (Avias, 1992).

During low-flow conditions, when the pumping rates exceed the natural discharge of the karst aquifer, the water level in the karst conduit and in the spring drops below the overflow level. Pumping then causes a drawdown of almost 30 m at the end of the low-water period, and the spring dries up (Fig. 4.1).

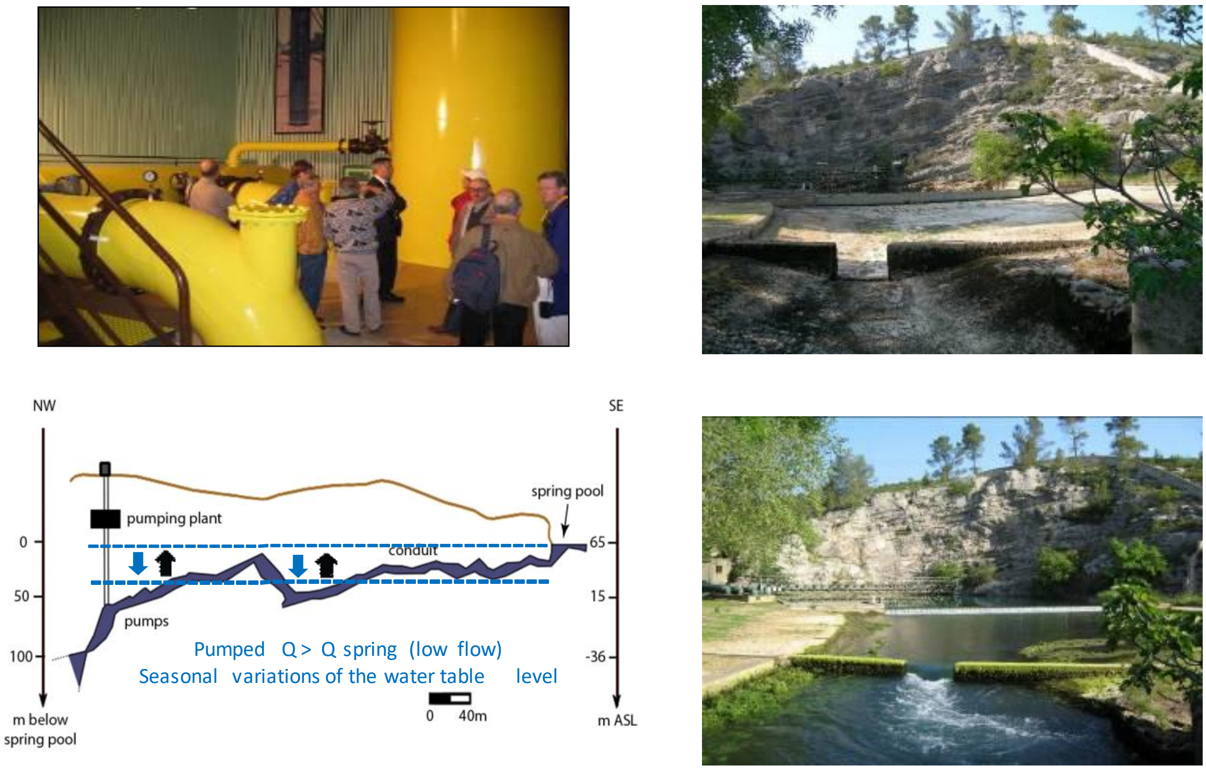

Figure 4.1: Lez spring during low flow and high flow conditions. Underground pumping station and seasonal variation of the water table level (Lez spring simplified topography from Mazzilli, 2011).

During autumn and winter, the karst aquifer is recharged and its reserves are renewed. The present water management scheme allows pumping at higher rates than the natural spring discharge during low-flow conditions, while supplying a minimum discharge rate into the Lez river for ecological concerns, and reducing flood hazards via rainfall storage in autumn.

The water quality of the Lez spring is good, except for bacteriological contaminations due to treated or not treated wastewaters infiltrating within the aquifer during flood events. After its subterranean pumping, water is supplied by pipes and treated at the Arago purification plant in Montpellier. Water is cleared of suspended particles by flocculation/decantation (if turbidity > 4 NTU) and disinfected with Cl_2 gas after filtering.

Since 2016, the Montpellier Mediterranean Metropole has the responsibility of the water resource management.

4.2 Water quality issues to justify the implementation of an EWS

The Lez spring is characterized by a complex mixing of mineralized waters with long residence times which derive from Jurassic and deeper aquifers, and recently infiltrated waters less mineralized and affected by anthropogenic contaminants (Bicalho et al., 2012). The different origins of the waters are: (1) superficial circulation within the main aquifer (Upper Jurassic and Cretaceous limestones), (2) surface-water interactions and interactions with the marls of the Valanginian cover of the aquifer, (3) water coming from deep circulation within Middle Jurassic and deeper compartments (Trias/Paleozoic) which can move up thanks to the major regional fault of Corconne-Les Matelles.

The proportions of these different water types vary during the hydrological cycle and depend on the hydrodynamical state of the aquifer. Previous studies showed that Lez spring waters have a good chemical quality even if they can be affected by punctual contaminations during flood events or very dry periods and may show peaks for faecal bacteria at these periods.

The study of land occupation and use provides information to clarify and quantify the anthropogenic activities on the Lez catchment. The major anthropogenic impacts on the water quality of the Lez spring are summarised below:

- Agricultural impacts

Agricultural activities correspond to approximately 25% of the surface area of the basin (Batiot et al., 2014). Vulnerable areas linked to these activities are essentially vineyards. These may explain the excess of the potability standards for phytosanitary products during high flows. Concerning agricultural contamination, regular determinations during a hydrological cycle (September 2010 to September 2011) of 16 pesticides in the waters of the Lez spring indicate a level of very low contamination (<25 ng/l). For some compounds, the concentration variations showed a seasonal use such as herbicides. These results have been compared with those resulting from others punctual samplings at the Lez spring (ADES data, 1997-2011). The average and maximum concentrations observed are generally low (respectively <30 ng/l and 50 ng/l) for all molecules. Punctually, compounds such as simazine or diuron may exceed 100 ng/l. As a result, the Lez aquifer does not appear to be chronically contaminated with pesticides, even if some molecules may exceed the potability standard set at 100 ng/l during flood events.

- Wastewaters impacts

In the Lez spring catchment, many areas have low permeable covers of low thickness, or present calcareous outcrops fractured and karstified which induce infiltration of water through the aquifer.

Urbanised areas represent about 5% of the basin. As other Mediterranean areas, the population of the basin doubled between 1990 and today. Wastewaters of the cities located in the southern part of the basin are collected and treated by the regional treatment plant of MAERA. In the northern part of the basin, urbanisation has sharply increased in some sectors, but infrastructures to treat urban or domestic wastewaters are note sufficient. So that, peaks of bacterial pathogen contents can be measured at the Lez spring during flood events or very dry conditions. The more vulnerable areas for water quality are sink-holes located in temporary streams where concentrated infiltration occurs during flood periods. Moreover, some are located near wastewater treatment plants which discharge effluents in these temporary streams, without dilution of the residual nutriments or bacterial contaminants after treatment. Due to their specific location around the major fault zone of Corconne-Les Matelles, the pollution is easily transported by subterranean flows to the Lez spring. So that, fast infiltration and pollution fluxes can be highlighted at the Lez spring by a decrease of Electrical Conductivity, and increase in bacterial compounds, TOC and natural fluorescence (Fig. 4.2).

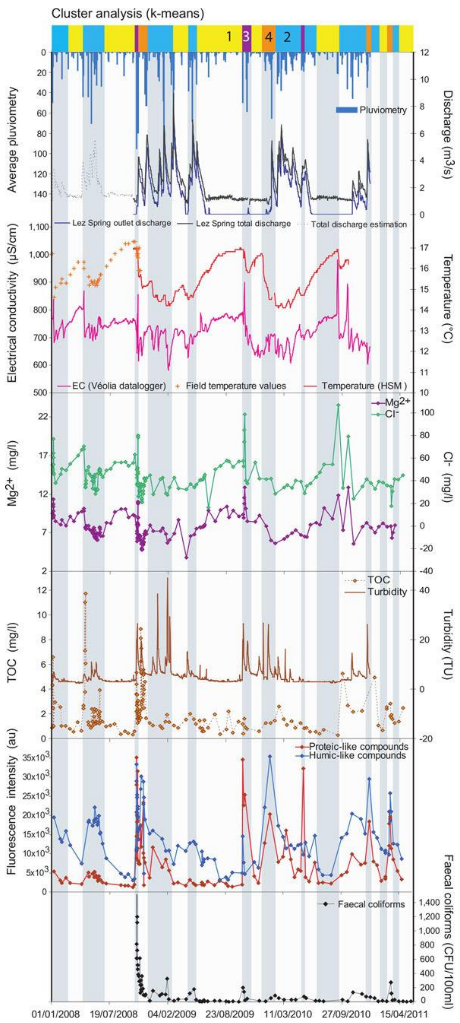

Figure 4.2: Temporal evolution of the fluorescence decomposed signal and the physico-chemical parameters measured between January 2008 and April 2011 at the Lez spring and the cluster analysis results (Quiers et al., 2014)

The fluorescence of Dissolved Organic Matter in karst systems is a suitable tool for tracing the origin and type of karst waters in addition to the more conventional hydrochemical parameters. The decomposition of the DOM fluorescence signal is a pertinent indicator for better analysing the total signal. Indeed, the signal emitted by the humic-like compounds is better appropriate for the monitoring of rapid infiltration flows during high-flow periods. The fluorescence of protein-like compounds provides further information on direct infiltration flows and their specific organic matter inputs. As the signal may be related to faecal bacteria, it highlights fast infiltration flows arriving at the outlet of the Lez spring, after few days, and demonstrates its vulnerability to contaminant flows, mainly related to domestic wastewater pollution.

4.3 Typology and monitoring set-up of the EWS

The study of karst springs is complex due to their high reactivity to flood events. To better characterize the aquifer functioning, regular and flood events water samplings need to be completed by in situ monitoring with high frequency measurements. Table 4.1 shows the different in situ monitoring parameters studied at the Lez spring. TOC, NO_3, NOM fluorescence, turbidity and 222-Rn can be used as EWS indicators to put in evidence fast infiltration within the aquifer and potential pollutions. Chloride may be relevant to monitor the contribution of waters coming from deeper compartments which are not impacted by anthropogenic pollution.

Data monitoring	Equipment specifications	Monitoring frequency	Time frame	Field regular sampling/measurement
Piezometry, EC, T°C	SDEC CTD probe	1 min	Ongoing from 2012	
T°C, pH, EC, DO, Cl	YSI6920 V2-2-SV probe	60 min	Ongoing from 2015	Twice a month + flood events
NOM fluorescence/turbidity	GGUN FL620	15 min	Ongoing from 2015	Twice a month for 3D fluorescence
TOC, DOC, NO3, Turbidity	Spectro ::lyser s ::can	15 min	Installation in November 2020. Calibration in progress	Twice a month + flood events for TOC, NO3
Radon-222	RAD7 Durridge	60 min	Ongoing from 2016	Twice a month + flood events

Table 4.1: Physico-chemical parameters measured in the field with high frequency monitoring which can be used as EWS indicators

4.4 First results: time series of selected chemical parameters recorded

Figure 4.3 shows the first results of natural fluorescence monitoring by in situ fluorometer (GGUN) compared to EEM fluorescence measured twice a month or more at the laboratory with a 3D spectrofluorometer (Erostate et al., 2016). Two types of organic compounds can be measured by in situ fluorimeter (AminoG/humic-like and proteic-like compounds), which correspond respectively to H1/H2 compounds (pedogenic origin) and P1 compounds (anthropogenic origin, Quiers et al., 2014). These monitorings are useful to identify fast infiltration within the system and contamination risk for the aquifer. The specific proteic optics of the in situ fluorimeter allows to highlight pollution peaks linked to wastewater pollution such as for the anomaly on 5/10/2015. Manual sampling carried out this day put in evidence a strong peak of fluorescence intensity for the proteic optics, related to an increase of Boron, Gadolinium anomaly, Faecal coliforms and Chloride contents, as well as a strong decreased in Dissolved Oxygen, which are typical indicators of a punctual contamination by waste-waters infiltration within the aquifer.

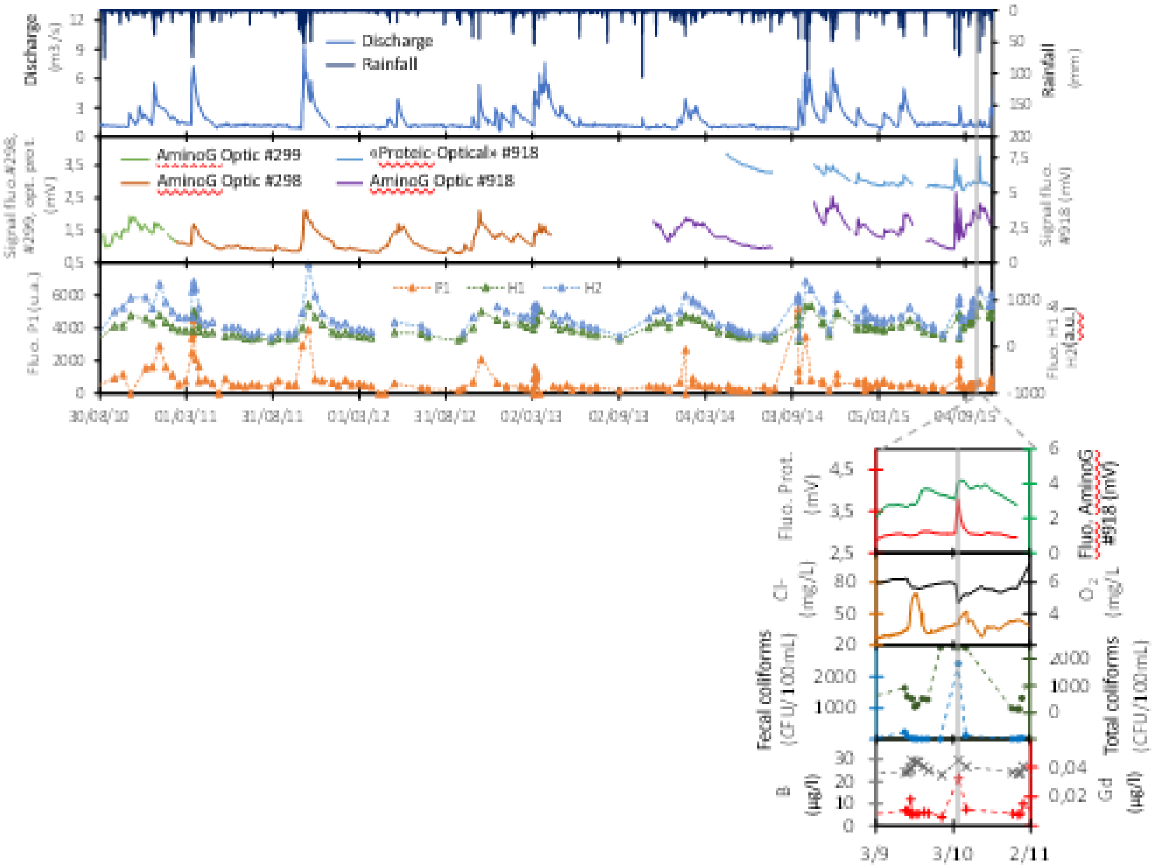

Figure 4.3: Temporal variations in (a) discharge, rainfall, fluorescence signal measured by in situ fluorometers and fluorescence measured in laboratory and (b) anomaly on 5/10/2015. (Erostate et al., 2016)

Figure 4.4 shows the time-series recorded at the Lez spring for different parameters which can be used as EWS indicators to follow the water quality. Regular samplings of chemical and/or bacterial parameters are indicated by symbols, whereas high frequency measurements achieved by in-situ monitoring are represented by lines (EC; turbidity; 222-Rn; natural fluorescence for AGA/humic-like and proteic-like compounds, Rohde, 2020).

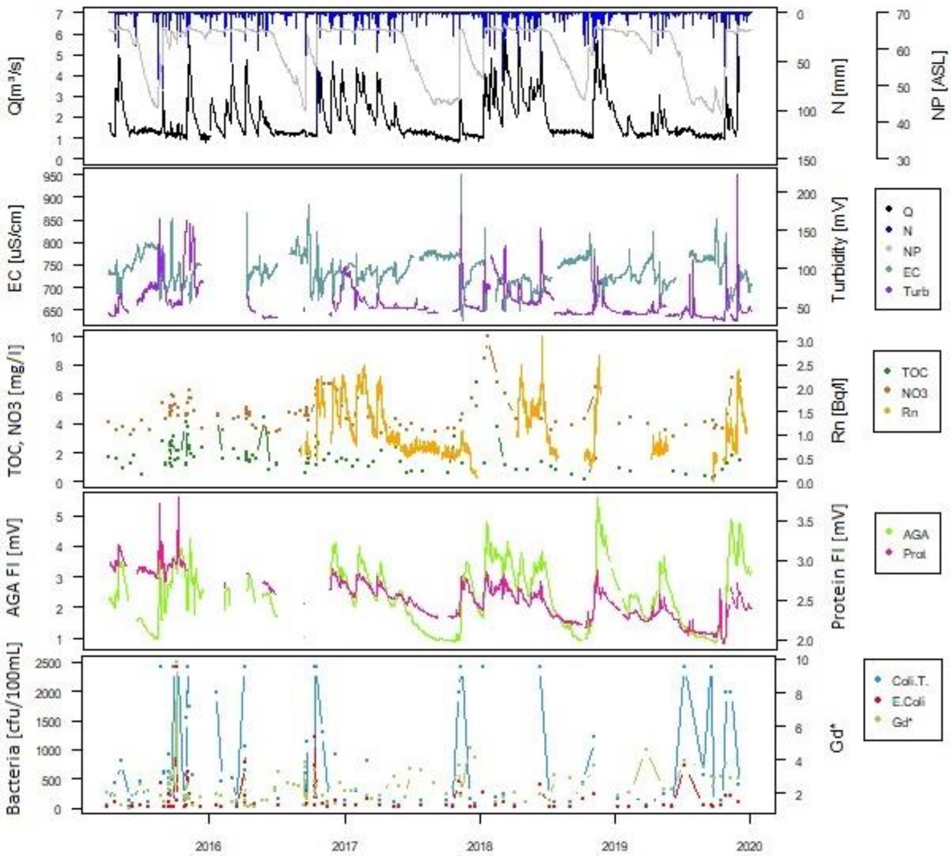

Figure 4.4: Temporal variations of different parameters (manual sampling and continuous monitoring) which can be used as EWS indicators for water quality at the Lez spring. (Rohde, 2020)

4.5 Data management and (statistical and/or numerical) modeling

Hydrological, hydrodynamical and hydrogeochemical data are collected and managed thanks to the data base of the MEDYCYSS Observatory (OSU OREME, OZCAR-THEIA, DEIMS-SDR eLTER):

https://data.oreme.org/observation/snokarst

https://deims.org/5b5ca767-0993-429c-8e58-a7efa39c936a

4.6 Operational benefits (expected) from the implementation of an EWS.

The *in situ* high frequency monitoring for the parameters presented in Table 4.1 has been validated by regular sampling and calibration of the different field probes/instruments. In the case of the Lez spring, we can monitor the influence of the more mineralised waters coming from deep compartments by continuous measurements of Chloride. Fast infiltration associated or not with punctual pollution can be put in evidence with TOC, NO3 and natural fluorescence in situ monitoring and make it possible to put in evidence pollution within the system and to specify its origin (wastewaters impacts for example). Indeed, in-situ parameters usually used to monitor water quality, such as turbidity, are not sufficient to indicate if there is an effective pollution or not linked to the recharge fluxes. The fluorescence monitoring of NOM with hydrochemical analysis is a complete set of tools that can characterise the recharge and the vulnerability of complex karst systems.

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5 Hochifen-Gottesacker karst area (test site in Austria)

5.1 Background

One objective within the framework of the KARMA project is to develop and implement improved monitoring tools for karst groundwater quality, and early-warning systems (EWS) for chemical and microbial spring water contamination at different time-scales, focusing on short-term contamination events, but also addressing long-term trends.

Karst springs are often characterized by long periods of sufficient water quality, interrupted by short but severe contamination events. It is a major challenge to identify these events in time and respond accordingly (Pronk et al. 2007). Under conditions of climate and land-use change, long-term trends in karst water quality are also a concern for many water suppliers, e.g. with respect to bacterial contamination, turbidity, nitrate and organic carbon.

The measurement of the enzymatic activity to determine bacterial contamination in near real-time with the ColiMinder was conducted in the Gottesacker area, Kleinwalsertal (Austria). This test site is ideal to evaluate the operational efficiency and reliability of the ColiMinder (proof-of-concept) because it is a highly variable and dynamic karst system and already well investigated. Therefore, this karst system is perfectly suitable to test and verify new and innovative methods and equipment within the framework of the KARMA project.

5.2 Field Site description

The test site is located in the Northern Calcareous Alps at the border between Austria (Vorarlberg) and Germany (Bavaria, Fig. 5.1a). The altitude varies between 1035 m asl (Sägebach Spring) and 2230 m asl (summit of Mt. Hochifen). The total size of the catchment area of Aubach- and Sägebach Spring (Fig. 5.1b) is about 35 km² (Chen & Goldscheider, 2014).

The study site belongs to the Helvetic zone, which plunges on three sides underneath the Flysch nappes consisting of marl and sandstone formations (Wyssling, 1986). The most important rock formation is the Cretaceous Schrattenkalk limestone layer, which forms the surface of the Gottesacker terrain (Goldscheider, 2005) and a relatively thin karst aquifer (about 100 m) above a thick marl formation (about 250 m) acting as a regional aquitard. Previous research (Goldscheider, 2005; Goeppert and Goldscheider, 2008) have shown that the orientation of the underground flow paths is structurally controlled, i.e. the underground flow is parallel to the strata.

The large but intermediate Aubach Spring discharges up to about 8 m³/s but runs dry in long dry periods and in winter. Further downstream, the Sägebach Spring presents the largest permanent spring in the valley and discharges up to about 3.5 m³/s (Chen & Goldscheider, 2014).

The mountain range SE of the Schwarzwasser valley is formed by sedimentary rocks of the Flysch zone and is mainly characterized by low permeability and drains by surface runoff. The karst aquifer in the catchment of the springs is recharged directly from precipitation, either diffuse as well as concentrated and also from surface streams that drain the part of the catchment area that consists of low permeable Flysch rocks (Chen & Goldscheider, 2014).

Both investigated springs, Aubach Spring (Fig. 5.2) and Sägebach Spring are high alpine karst springs and currently not used for drinking water supply, therefore the water is not treated in any way. The base-flow spring (Sägebach Spring) is partly used for a hydropower plant.

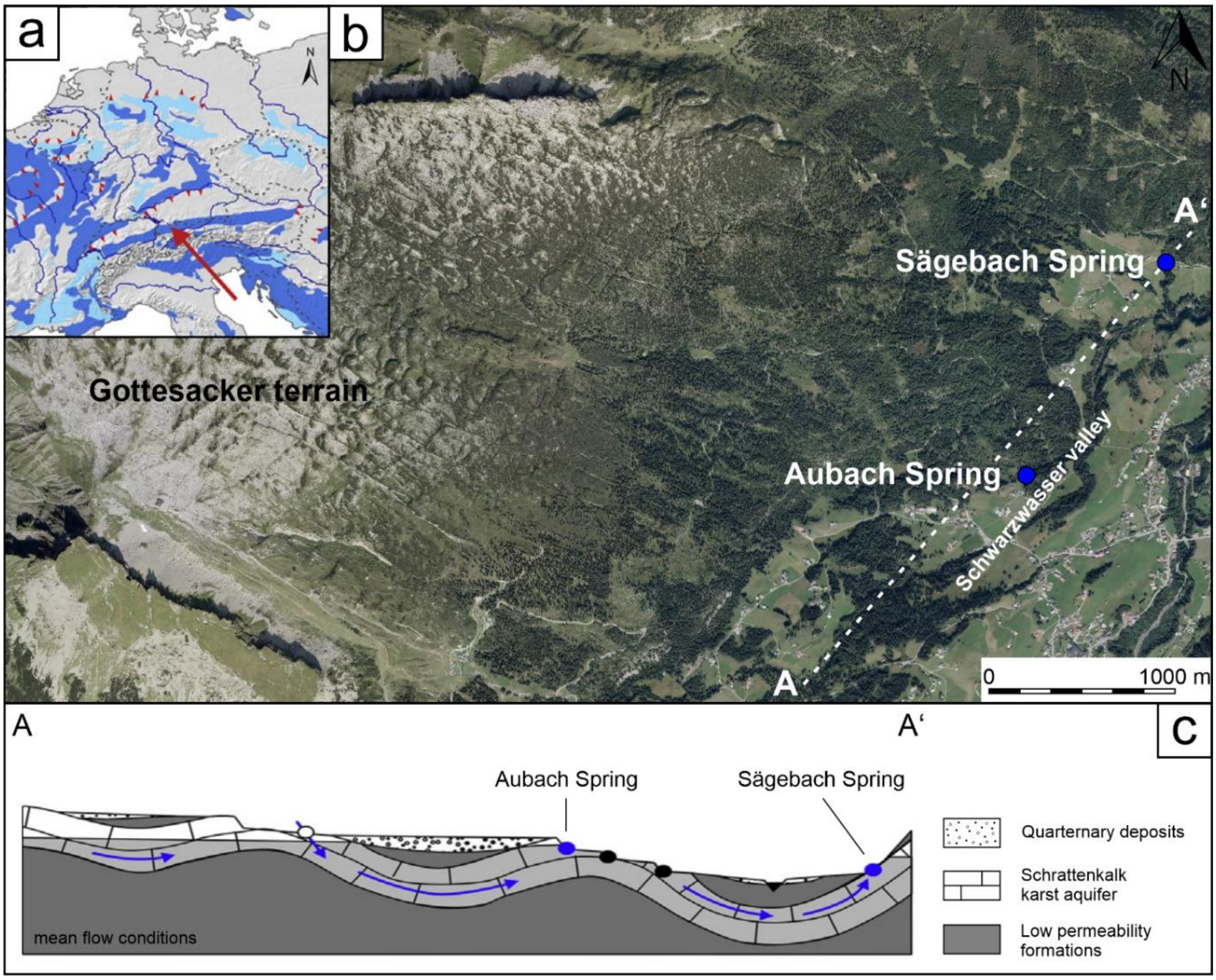

Figure 5.1: a) Location of the test site shown on a section of the World Karst Aquifer Map (Chen et al., 2017) with carbonate rocks in blue. b) Detail of the test site with the Gottesacker area and Aubach- and Sägebach Spring (basemap: Land Vorarlberg – data.vorarlberg.gv.at) and c) schematic cross-section with flow paths at mean flow conditions (modified after Goeppert et al., 2020).

Both springs show a high variability regarding the water quality. After rain events, high contaminations with fecal bacteria, caused by livestock farming and wild live, can be observed. In addition, the water quality of Aubach Spring is influenced by an old sewage pit. Industrial pollution, pesticides, and herbicides do not play a role, since both springs are alpine karst springs and there is no intensive land use in the Kleinwalsertal and especially not in the catchment area of the two springs.

Figure 5.2: Picture of Aubach Spring.

Figure 5.3: Picture of Sägebach Spring with the installed equipment.

5.3 Material and methods

In addition to the standard parameters discharge, electrical conductivity, water temperature, pH and redox, the particle load and particle size-distribution (PSD) and the natural fluorescence were measured. The PSD was measured with the Klotz particle counter PCSS fluid lite and the natural fluorescence was measured with an Albillia field fluorometer FL30. *E.coli* bacteria were determined using a conventional culture-based method (IDEXX ColiSure) and with a novel instrument (ColiMinder) based on enzymatic activity measurements. Manual water samples were taken to determine the chemical composition of the spring water and total organic carbon as well as excitation-emission matrices (EEMs) using a laboratory Aqualog fluorometer. The setup of all instruments at Sägebach Spring is shown on Fig. 5.3.

PCSS fluid lite:

The particle counter gives the number of suspended particles of 16 different definable size classes in a range of 0.9 to 150 µm. A particle passes a laser beam, which results in a decrease in laser light in the detector. The extent of the decrease is given by assuming a spherical shape for the diameter of the particle. In this study, each measurement was started with a rinsing cycle, where 40 ml sample water ran through the device, followed by the measurement cycle where 10 ml sample water were used to determine the particle load and the respective size distribution. The following size-classes (in µm) were predefined and measured: 1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 15, 20, 25, 50, 75, 100.

In this study, especially small particle size classes were considered, since pathogenic bacteria are in the range of ca. 1-3 µm (Goeppert and Goldscheider, 2011; Reshes et al., 2008). Furthermore, small particles have a higher mobility and can be transported through the whole aquifer.

FL30 field fluorometer:

In this study, two GGUN-FL30 field fluorometers were used, one at Sägebach Spring and one at Aubach Spring. The FL30 consists of a flow-through measurement cell, which is placed directly in the spring water, and a data logger. The logging interval can be set manually between 2 seconds and 15 minutes. The used field fluorometers are equipped with optics for uranine, tryptophan and amino G acid. The GGUN-Fl30 also measures the turbidity.

ColiMinder Mobile:

The ColiMinder measures the enzymatic activity of certain enzymes, in our case β-glucoronidase (GLUC, *E. coli*). The measurement is carried out in the liquid phase and a single full working cycle takes approx. 30 minutes including cleaning. One measurement is based on an increase in fluorescence intensity, resulting from the activity of GLUC and, hence, the accumulation of the highly fluorescent reaction product 4-Methylumbelliferone (Ender et al., 2017). For a single determination, about 10 to 15 ml of sample water is required. Briefly, for each measurement, fresh unfiltered water is pumped into the measurement chamber, where it is mixed with a defined buffer and the fluorogenic substrate solution. Since maximal GLUC activity can be observed at 44 °C (George et al., 2000), the solution is preheated to a constant temperature of 44.0 ± 0.1 °C, before the measurement process is started automatically. The rate of enzymatic reaction is determined in volts per second. In certain intervals, blank measurements with clean deionized water as a sample are performed.

Measurement intervals can be set up manually with a minimum of 30 minutes. Our measurements were mostly done with one to four hour intervals.

5.4 Preliminary Results

Aubach Spring:

The first results of the standard parameters of our measurement campaign at Aubach Spring are given in Figure 5.4. Noticeable are the large discharge variations between 150 l/s and over 6000 l/s. Fig. 5.4 also shows the almost immediate reaction of discharge, electrical conductivity (EC) and temperature after rainfall events. Depending on the intensity of rainfall, the EC decreases between 30 and 60 µS/cm. Accordingly, water temperature decreases between 0.4 and 0.8 °C. An exception is the rain event on 17th and 18th of August where temperature increases by 0.5 °C, probably related to rainfall mainly in the direct surrounding of Aubach Spring, which would also explain the higher turbidity compared to other events (Frank et al., in preparation).

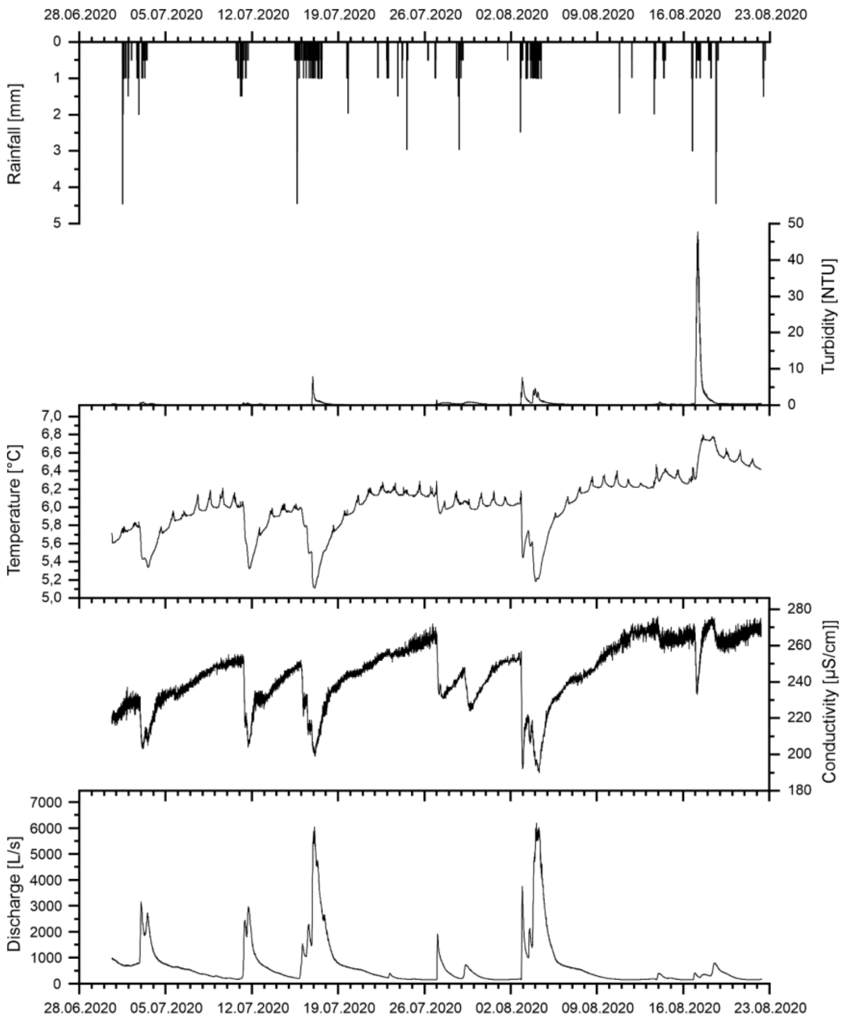

Figure 5.4: Time series from Aubach Spring, including discharge, electrical conductivity, temperature and turbidity, together with the rainfall data from a nearby station (Frank et al., in preparation).

Time series of the major ions Ca^{2+}, Mg^{2+}, HCO_3^- and SO_4^{2-} are given in Figure 5.5. Water samples were taken in two to eight hour intervals and analyzed in the laboratory with an ICP-MS (Agilent Technologies 7800) and an IC (Dionex ICS-1100). The water chemistry also shows a fast and marked reaction to increasing discharge as can be seen especially after the rain events on 16th of July and 3rd of August where all major ion concentrations decrease. Bicarbonate values drop by 30 mg/l (ca. 22 %), sulfate by 3.5 mg/l (ca. 85 %), calcium by 8 mg/l (ca. 20 %) and magnesium by 2 mg/l (ca. 80 %).

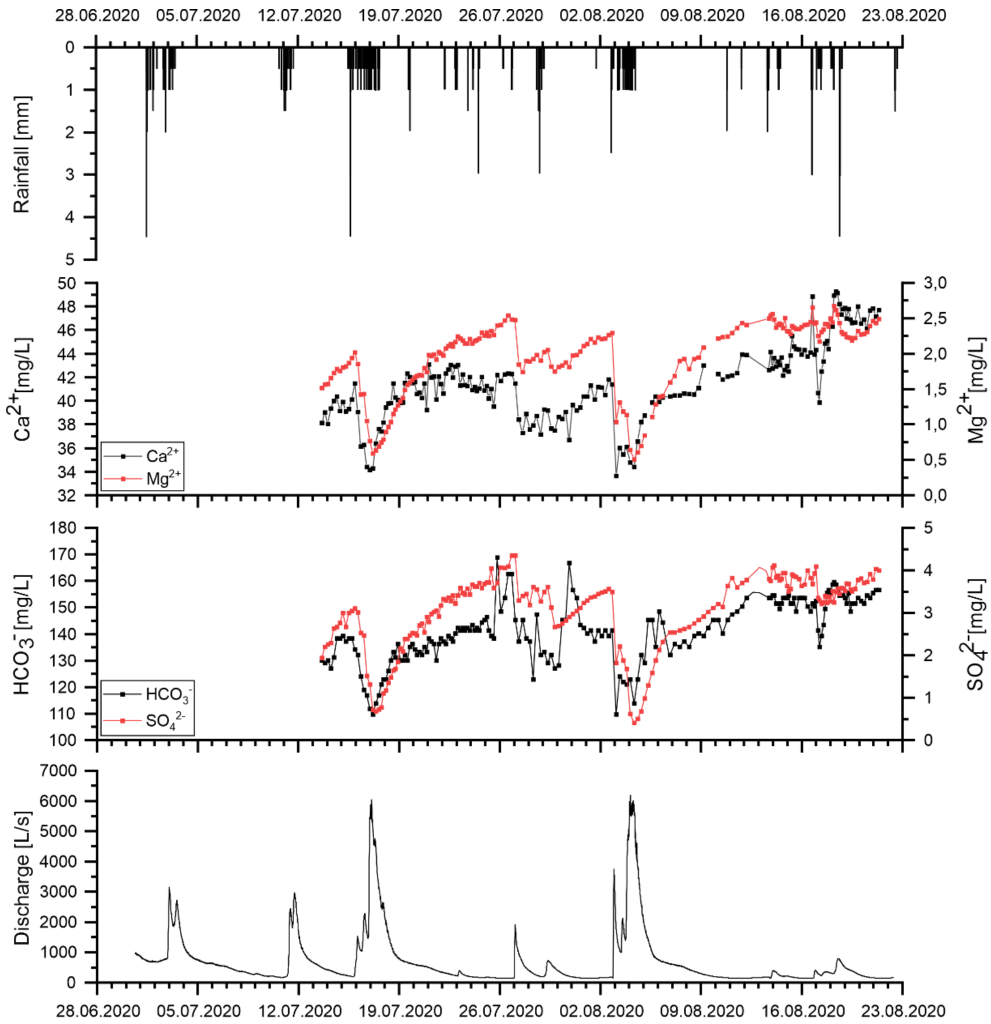

Figure 5.5: Time series from Aubach spring with the major cations (Calcium, Magnesium) and anions (Bicarbonate, Sulfate), together with discharge and rainfall.

Sägebach Spring:

A major focus was the enzymatic activity, which can be measured with the ColiMinder. In addition, a conventional culture based method was used to determine the fecal indicator bacteria *E. coli*. Figure 5.6 shows the enzymatic activity, together with the determined E. coli values with the IDEXX method and with the recorded turbidity after a rain event on 17th of August. A clear correlation between the enzymatic activity and the *E. coli* values was detected. The enzymatic activity increased from 3 to 13.5 and the E. coli values by ca. 900. Turbidity also showed a marked increase from almost zero to over 50 NTU (Frank et al., in preparation).

Figure 5.7 shows the particle load and the particle size-distribution measured after a rain event on 23rd and 24th of July at Sägebach Spring. All particle size classes, except the 100 µm class, show a sharp increase with a clear peak after the rain event. The smaller size classes (1 µm to 8 µm) show a secondary peak, which can be attributed to the arrival of allochthonous material from the land surface (Goldscheider et al., 2010; Frank et al., 2018). The larger size classes (9 µm to 75 µm) only show one peak, while the largest particles measured in this study (100 µm) do not show a clear peak at all, most probably because of sedimentation (Frank et al., in preparation).

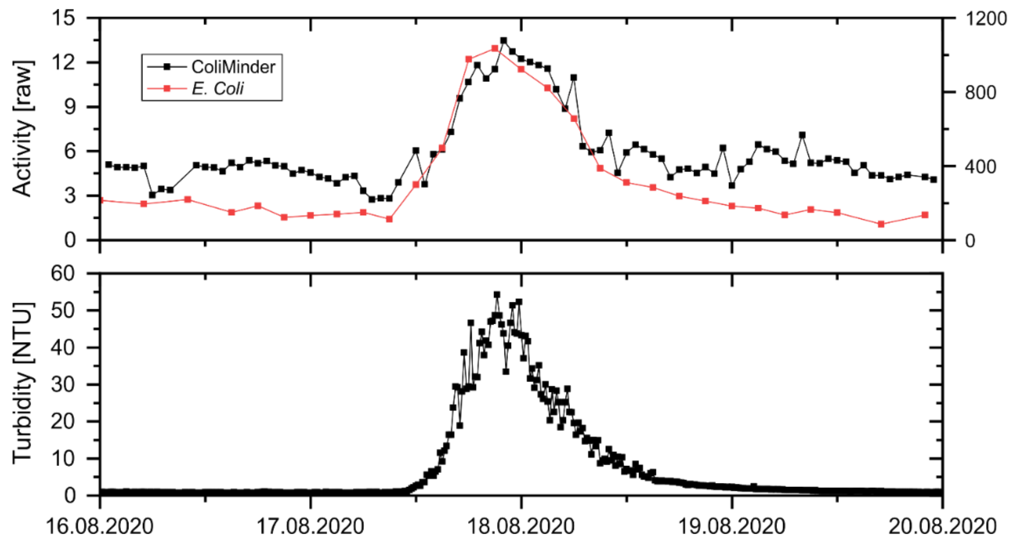

Figure 5.6: Measured enzymatic activity [raw] with the ColiMinder, E. coli values in MPN/100ml, determined with the IDEXX ColiSure method and the measured turbidity (Frank et al., in preparation).

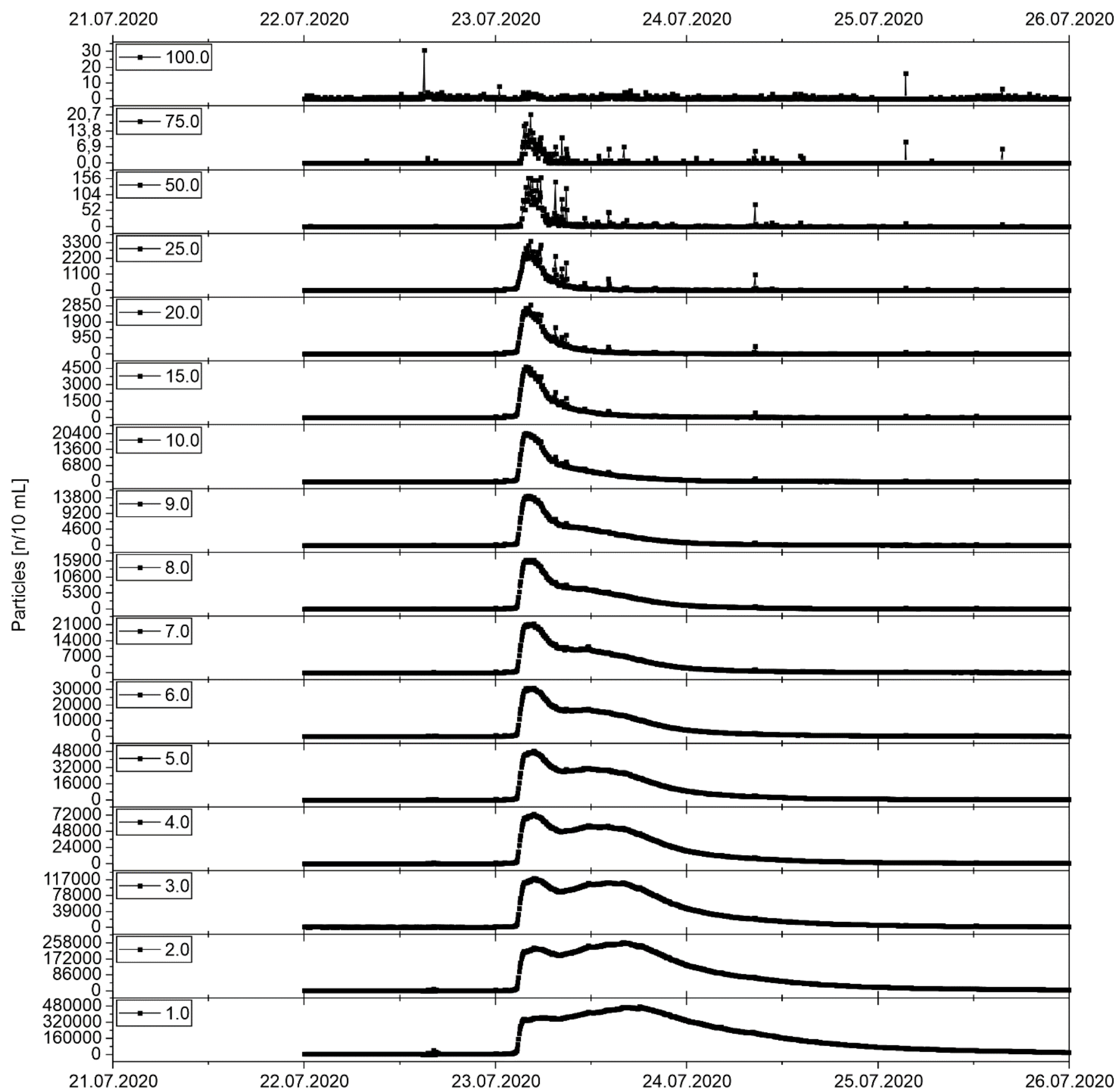

Figure 5.7: Particle load (n/10 ml) of all 16 measured particle size classes at Sägebach Spring (Frank et al., in preparation).

5.5 Application of an Early Warning System

An important element in the control of water quality is the detection of contaminants in the water prior to its use as drinking water. Early warning systems can help to identify critical contaminants and help to initiate counteractions before the distribution of the water.

In our test site, we could prove different methods and identify several parameters that are potentially suitable for an early warning system. The results of the small particle size-classes at Aubach Spring clearly showed two identifiable peaks from which the second one can be attributed to the arrival of surface water entering the karst aquifer. This second peak also correlates with higher values of the fecal indicator bacteria *E. coli*. Therefore, the small particles could be used as an early warning system for bacterial contamination at Aubach Spring.

At Sägebach Spring we demonstrated that the measurement of the enzymatic activity correlates with conventional determination methods of *E. coli* and also with turbidity measurements. This means that at Sägebach Spring the parameters enzymatic activity and turbidity could be used as a real-time indication system for bacterial contamination.

5.6 References

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6 Conclusions and outlook

Selected experimental sites within KARMA project (Lebanon, Spain, France and Germany/Austria) have a strong background hydrogeological knowledge and particular spring water contamination related-issues, which make them ideal for the development and implementation of an Early Warning System facing karst groundwater resources protection.

Generally, in each test site numerous regional and specific hydrogeological investigations have been performed during the last years aiming a better understanding of the karst system functioning. In all of them, the contamination episodes investigated are associated with urban wastewater leakages (point source pollution) and agricultural and industrial activities (diffuse pollution); in which surface waters act as transport media for the latter infiltration of the contaminants through sinking points/stretches over karst exposures.

Common measurement protocols for groundwater monitoring have been established in the four experimental sites and the application of fluorescence-based techniques, particle size distribution (PSD) analysis and microbiological methods, which constitute an ideal configuration of sensors that generally fits well to the typologies of water pollution episodes previously described. Additionally, complementary and specific measurements of chemical, isotopic (stable and radioactive) and microbiological parameters, more adapted to the background of the groundwater quality status in each site, provide further hydrogeological information that will serve to better characterize transport processes of contaminant along groundwater flowpaths. This common measurement strategy will ensure the exchange of field experiences among project partners, but also the definition of the monitoring setup employing the minimum number of specific parameters/sensors to gather the maximum information for forecasting the arrival of polluted groundwater from other basic ones (electrical conductivity, turbidity, water temperature, etc.).

After the execution of the first development phase of the task 3.2, the following goals have been successfully achieved in each KARMA test site: a) hydrogeological characterization and karst aquifer functioning; b) assessment of pollution sources; c) establishment of the monitoring network and; d) selection of the set of chemical/bacteriological/isotopic indicators for a precise characterization of groundwater contamination events at both, short and long term.

Next research advances must be focused on the refinement of individual EWS implementation protocols as described in Marín et al. (2020), which need to be properly adapted to each test site according to their hydrogeological characteristics (aquifer system) and configuration of drinking water production network (hydraulic facilities). Among the critical phases for EWS implementation, automatic and remote data collection need to be addressed by exploring different telemetry options (e.g. *ad-hoc* systems, commercial solutions, etc.). This will facilitate the communication with field devices and will allow for taking technical decisions to the water operators in case of the probable occurrence of a contamination episode. The ultimate challenging issue will be to get a unique *on-line* water quality monitoring technology, by integrating a telemetry system with an open software including statistical and numerical algorithms for launching warning messages to the water operators. Depending on the final evolution of each EWS, this could also include the automatic control of simple hydraulic features (valves, pipes, etc.) by which the drinking water management be also improved in a more technical way. Then, the acquired scientific knowledge will be fully transferred into the drinking water system functioning for the benefit of the end-user.