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Research paper

Rooftop rainwater harvesting by a shallow well – Impacts and potential from a field experiment in the Danube-Tisza Interfluve, Hungary

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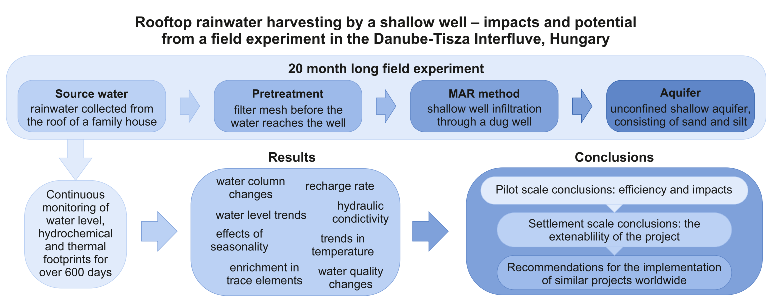
HIGHLIGHTS

- A total of 60.616 m^3 of rainwater was infiltrated in a 20-months-long pilot project.
- Groundwater level, temperature, hydrochemical and isotopic changes were monitored.
- Major water quality improvement was achieved and maintained in the infiltration well.
- Elevated zinc concentrations were identified in roof-runoff and groundwater.
- Water level time series analysis indicated clogging and the need for maintenance.

GRAPHICAL ABSTRACT



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ABSTRACT

Groundwater levels have declined significantly in the last decades in many parts of the world, due to anthropogenic activities and climate change. The study focuses on the long-term potential and environmental impacts of rooftop rainwater harvesting coupled with shallow well infiltration, which is a local scale inexpensive solution that could contribute to easing water shortage. The Danube-Tisza Interfluve (Hungary) was used as a study area, where a field experiment was set up, funneling rainwater from the roof of a family house to the shallow dug well in the yard, connected to a porous unconfined aquifer. Changes in groundwater levels, as well as thermal, hydrochemical and isotopic footprints were monitored and evaluated for over 600 days to determine the quantitative and qualitative effects of this method and to identify the long-term physico-chemical impacts of infiltrated water on ambient groundwater. During the monitoring period, 60.616 m^3 of precipitation was infiltrated through the shallow well that could achieve significant water quality improvement: Mg^{2+}, Na^+, Cl^-, SO_4^{2-}, NO_3^- concentrations and TDS decreased by 35–88% after the infiltration started and a further 35–96% decrease

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occurred in these parameters by the end of the monitoring period. Rooftop-collected water was enriched in Zn, Sr, Cu, Mn, Ba and Al, their concentrations being 1.9–48.2 times higher, in roof runoff than in the precipitation. The monitoring of water column changes and infiltration curve analysis after precipitation events helped identifying the clogging process that decreased the hydraulic conductivity of the well bottom by one order of magnitude. The results of this research provide information on the efficiency and environmental impacts of the pilot project and contribute to the extension of the design to town level and to similar settlements in the area. Additionally, general conclusions can be drawn to promote the implementation of similar projects in porous unconfined aquifers worldwide.

1. Introduction and aims

Due to climate change, weather conditions are becoming more and more extreme, with longer periods of drought and flood causing environmental, agricultural and consequent health, infrastructural, social and economic problems worldwide (Treidel et al., 2011; Taylor et al., 2013; Harrison et al., 2015; Arnell et al., 2016). At the same time, the need for irrigation increases the use of fresh groundwater globally (Zhou et al., 2010; Wada et al., 2013; Wu et al., 2020). These two effects can cause serious water shortage in many parts of the world including e.g., California (USA), northern India, the North China Plain, the Middle East (Rodell et al., 2018) etc.

In the field of water management, one of the most significant ways to achieve adaptation to climate change is MAR, which is the purposeful recharge of water to aquifers for subsequent recovery or environmental benefit (NRMMC, 2009). It is a group of nature-inspired approaches that promote the accumulation of water reserves under the surface and support the joint management of surface waters and groundwaters (Evans and Dillon, 2018; Dillon et al., 2018). Managed Aquifer Recharge is a suitable way to reduce the inequalities of water conditions and helps mitigating the related consequences. This solution can contribute to the rehabilitation of groundwater depletion, restoration of pressure levels in aquifers, avoiding declining yields, halting ground subsidence, improving water quality and maintaining groundwater-dependent ecosystems (e.g., Gale, 2005; Dillon et al., 2009; Dillon et al., 2018). Different types of MAR methods can be used, based on the local environmental conditions and opportunities (geomorphology, geology, water availability, hydrogeology etc.) as well as on the aims of the aquifer recharge (IGRAC, 2007; Dillon et al., 2009). Spreading methods (e.g., infiltration ponds and basins), induced bank filtration, and well, shaft and borehole recharge are used to enhance infiltration and replenishment, while in-channel modifications and rainwater and runoff harvesting are mainly aimed at intercepting the water (IGRAC, 2007). These methods can be well combined with each other, e.g., rainwater and runoff harvesting can serve as the water source for infiltration basins or wells to replenish groundwater reservoirs. Rainwater can be infiltrated via ponds, basins and ditches through the unsaturated zone or using wells thus leading water directly to the saturated zone (Gale, 2005; Dillon et al., 2009). The latter solution has several advantages, including lower evaporation loss, faster increase of groundwater level, smaller surface area use, and lower risk of biological activity. On the other hand, adequate water quality of infiltrated water is crucial in this case, in order to avoid groundwater contamination (Gale, 2005; Dillon et al., 2009; Casanova et al., 2016; Page et al., 2018).

One of the areas in Hungary suffering from serious water shortage is the Danube-Tisza Interfluve (DTI; Fig. 1), where water level reduction and related water management problems started in the 1970s (Major and Neppel, 1988). The water shortage of the area can be attributed to several factors. Climate change, especially the reduction and change in the distribution of precipitation and yearly average temperature increase played an important role in the occurring processes. Additionally, several types of human activities, such as land use changes, afforestation, shallow and deep groundwater abstraction, canalization and land drainage contributed to the water level decrease (Pálfai, 1993, 2010; Kovács et al., 2017). The degree of water level decline is approx. 2–3 m, but in some areas, it can reach even 6–7 m (Major and Neppel, 1988; Szilágyi and Vorosmarty, 1997; Szalai and Nagy, 2006). These processes caused major agricultural, ecological and other related social problems (Ladányi et al., 2009; Kovács et al., 2017). In the ridge part of the DTI, many shallow wells and lakes have dried out. As an example, Lake Kondor (SW of Kerekegyháza, Fig. 1c) as a wetland almost entirely disappeared (Ujházy and Biró, 2013), and most of the dug wells of Kerekegyháza run out of water (based on personal communications with the residents).

Many plans were worked out in the last decades, involving large, cross-regional technical investments, but none of them fully materialized due to financial and ecological concerns (Kovács et al., 2017). Different replenishment scenarios were made for channeling river water from the Danube to the area (e.g., Orlóci, 2003; Alföldi and Kapolyi, 2011; Nagy et al., 2016). Other plans tried to offer a solution by constructing reservoirs, even using them for infiltration (Nemere, 1994; Gyirán, 2009; Nagy et al., 2016).

The problem has been unresolved for decades, but its severity is growing in light of the ongoing climate change, as DTI is one of the most vulnerable areas in the country in this regard (NCCS, 2018). In Hungary, the annual average temperature is expected to increase by 4–5.4 °C and the annual precipitation is likely to decrease by about 20% by the end of the century (Pieczka et al., 2011). A further increase in weather extremes (e.g., occurrence of droughts) are expected (Bartholy et al., 2014), as well as a significant decrease in the climatic water balance (Rotárné Szalkai et al., 2015).

The unconfined aquifers have adequate storage capacity, and this would provide an opportunity for more efficient water retention and water replenishment on local scale (Páris, 2009), Therefore, one of the aims of this research is to evaluate the water replenishment possibilities of the shallow porous aquifers of the DTI from a different perspective by investigating a local water replenishment method. Rooftop rainwater harvesting (RRWH) coupled with shallow well infiltration (RRWH-SW) was selected for this purpose for the following reasons: i) The only possible local source of replenishment in the area is rainwater. ii) Using dug wells is possibly the most inexpensive and most convenient way of water replenishment in the area. These wells are already available and usually not used anymore because either they have dried out, or the water quality of shallow groundwater is not adequate anymore. By using them for infiltration, they could have a new purpose in water management. iii) Using dug wells for rainwater collection or disposal is an existing practice in the area, however it is not favored by the authorities as it might pose a risk to groundwater quality and fine sediments may clog the pores of the well bottom (Ministry of Interior, 2017). In order to recommend these systems to the authorities and residents, assurance of the effectiveness of the method is needed, and the possible downsides should be assessed. Consequently, the second aim is to demonstrate the quantitative and qualitative potential and environmental impacts of the method.

Several former research proved that rainwater harvesting, either collected from roofs and roads can be an effective tool to increase groundwater levels, especially on catchment scale (e.g., Sayana et al., 2010; Glendenning and Vervoort, 2010; Jebamalar et al., 2012; Jebamalar et al., 2021). Mapping of suitable locations for rainwater and runoff harvesting is a prevalent research objective (e.g., Mati et al.,

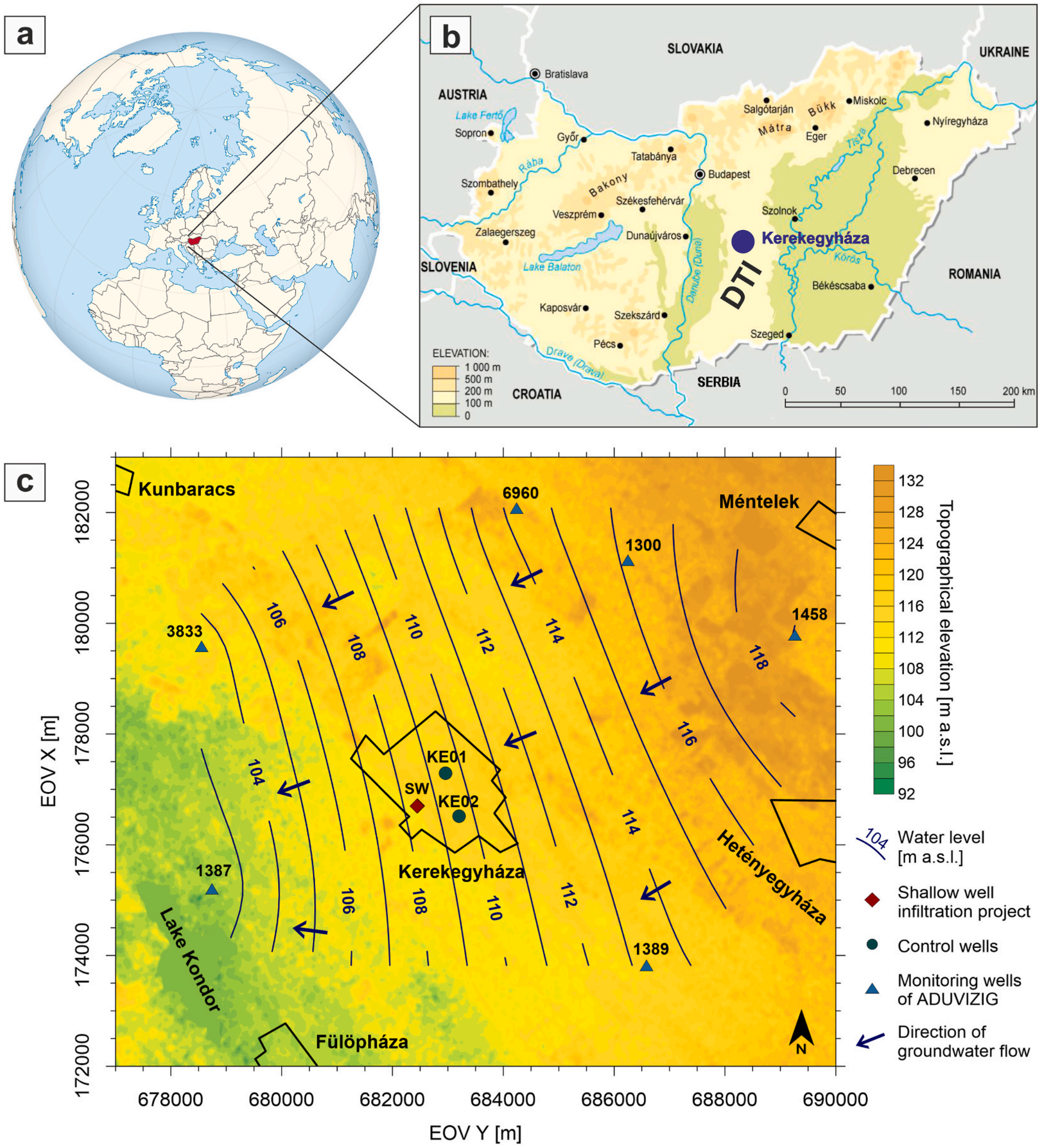

Fig. 1. Location of the study area: (a) Hungary, (b) Danube-Tisza Interfluve (DTI) and Kerekegyháza, (c) shallow well infiltration project, control wells and other monitoring wells in the area. Note: The map of Fig. 1c uses the Hungarian EOV grid (Uniform National Projection) in which EOV X represents northing and EOV Y represents easting in meters. The digital elevation model (resolution: 5 × 5 m) was acquired from the Lechner Knowledge Center. The topographical elevation is referenced to the Baltic Sea level. The water level contours were constructed based on data measured in the monitoring wells by ADUVÍZIG at the beginning of the project.

2006; Ramakrishnan et al., 2009; Kadam et al., 2012; Adham et al., 2018; Jasrotia et al., 2019; Shyam et al., 2021) to promote the implementation of new MAR systems and help decision-makers. Although rooftop rainwater harvesting is getting more and more popular and several research deal with planning of such systems (e.g., Dwivedi et al., 2013; Nachshon et al., 2016; Farswan et al., 2019; Siddiqui and Siddiqui, 2019; Diwan and Karanam, 2020; Gado and El-Agha, 2020; Mishra et al., 2020), only a few case studies focus on the assessment of the performance of local scale rooftop rainwater harvesting systems combined with wells (e.g., Dillon and Barry, 2005; Venugopal and Ghosh, 2010; Jebamalar and Ravikumar, 2011; Barry et al., 2013; Pawar et al., 2014; Wang et al., 2015; Hasan et al., 2018; Pavelic et al., 2020; Rahaman et al., 2019a,b).

The scale of these studies varies between individual system (e.g., Venugopal and Ghosh, 2010) and neighborhood or settlement level (Jebamalar and Ravikumar, 2011; Pawar et al., 2014). Monitoring of individual systems, based on field experiments varies in length and frequency as well. Dillon and Barry (2005) had a 2 year long monitoring scheme of measuring water level, EC, T, injected volume and turbidity during infiltration to a borewell. Barry et al. (2013) analyzed clogging related to this pilot. Venugopal and Ghosh (2010) had quite a long monitoring of 7 years, but they only measured water level seasonally, no chemical measurements have been made. Wang et al. (2015) analyzed rainfall amount, water level, main ions, metals, metalloids and nutrients for 6 months in a deep (~230 m) karstic aquifer but this is not applicable in shallow porous environments. Hasan et al. (2018) monitored EC, temperature, pH, turbidity, water level, infiltrated water amount, As and Fe concentrations for 5 months and they achieved a significant water quality improvement. Pavelic et al. (2020) studied 5 MAR trials where they collected rainwater from rooftops, unpaved roads and local fields. Their monitoring scheme lasted for 2 hydrologic years: rainfall amount, groundwater levels and temperature were measured by automatic devices; general inorganics, metals, nutrients, microbial pathogens and pesticides were measured seasonally. Rahaman et al. (2019b) only had

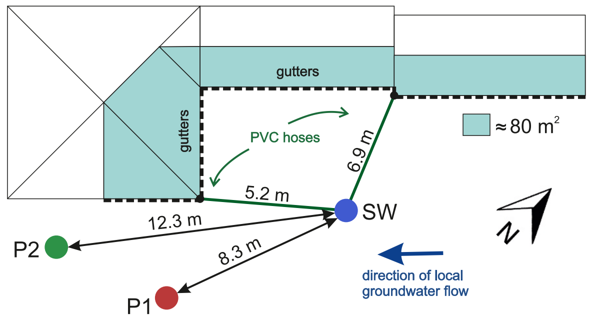

Fig. 2. Schematic top view of the pilot site indicating the location of the wells (SW, P1, P2), gutters and PVC hoses, the used area for water harvesting and the direction of local groundwater flow.

pre- and post-MAR measurements of water quality. None of the above-mentioned case studies analyzed clogging of the wells, except for Barry et al. (2013). From these studies focusing on individual systems, only a part of them used shallow dug well for infiltration (Venugopal and Ghosh, 2010; Pavelic et al., 2020; Rahaman et al., 2019a,b), and from these only Rahaman et al. (2019a,b) infiltrated into a porous aquifer.

Although, rooftop rainwater is generally considered as rarely polluted (Meera and Mansoor Ahammed, 2006), its quality depends on several important factors, like pollutant concentration in the rain, location of the site (distance from pollutant sources), material and physical properties of the roof and gutters, among others (Förster, 1999; Abbasi and Abbasi, 2011; Meera and Mansoor Ahammed, 2018). In case of metal roofs and metal gutters, Zn, Cu and Pb are common heavy metals in the harvested rainwater (Meera and Mansoor Ahammed, 2006). However, for tile roofs, the enrichment factor of Zn in roof-runoff is much lower. Other parameters, such as turbidity, hardness and nitrate content can increase in roof-runoff from tile roofs (Meera and Mansoor Ahammed, 2018).

In order to guarantee the long-term efficiency of RRWH-SW systems in a porous aquifer, both in terms of quantity and water quality, the implementation of a field experiment seems to be an adequate method. However, in order to exclude the effects of seasonality, a continuous monitoring scheme and an observation period longer than one hydrologic year is needed.

Based on these preliminary considerations, the objectives of this study were to carry out a long-term experimental field study i) to assess the quantitative and (ii) qualitative potential of rooftop rainwater harvesting coupled with shallow well infiltration (RRWH-SW); iii) to determine the related environmental consequences (especially related to groundwater contamination); iv) to examine the effect of clogging; and v) to evaluate the significance of the results on settlement-regional scales and globally. Although the results of the experimental study directly widen the scope of water replenishment possibilities in the DTI, the results for an unconfined porous environment can also provide thorough information on the solution's long-term potential and environmental impacts.

2. Characterization of the pilot site in the study area

The study area is located in the Danube-Tisza Interfluve area, Hungary (Fig. 1). The DTI is a ridge region, up to 130–140 m a.s.l. Between Danube and Tisza Rivers. The river valleys are situated at 85–90 m above sea level. The experiment is located in a small rural town, Kerekegyháza, which is situated on the western side of the elevated ridge, on the catchment area of the Danube. In the surroundings of Kerekegyháza, the topographical elevation difference is less than 10 m (109–118 m a.s.l.; Fig. 1c).

The near surface aquifers and aquitards of the area consist of sand, coarse silt, peat, mud, clay and calcareous mud. The shallow groundwater flow systems of the elevated ridge region are under the effect of topography-driven meteoric flow regime and in shallow depth $Ca,Mg(HCO_3)_2$ type waters are predominant (Mádl-Szőnyi and Tóth, 2009). The study area is located in the recharge area, the vertical hydraulic gradient is approx. $2–4\bullet10^{-2}$ (Mádl-Szőnyi and Tóth, 2009). Based on Fig. 1c, the horizontal hydraulic gradient is one order of magnitude lower ($\sim1.5\bullet10^{-3}$).

The climate of the study area is moderately warm and dry. The average annual temperature is around 11 °C. The annual average rainfall is 500–550 mm y^{-1} with an uneven temporal distribution. The number of hours of sunshine is considered high in Hungary, exceeding 2000 h per year (OMSZ website). The rate of evapotranspiration is 80–90% of the annual precipitation, on average 470 mm (Szilágyi et al., 2012). Using $^3H/^3He$ age profiling to a well nest in a near study area (~5 km), the recharge rate has been estimated to be 48 ± 6 mm y^{-1} (Palcsu et al., 2017), which is about 9% of the annual precipitation being in a good agreement with the evapotranspiration rate.

In January 2020, a field experiment was set up funneling rainwater from the roof of a family house into an abandoned shallow dug well (SW) located in the house yard (Fig. 2). SW is 6.2 m deep with an open bottom, its diameter being 0.8 m. The well is screened in an unconfined shallow aquifer consisting of sand and silt. Although SW was not used in the past, we found it in excellent conditions; the concrete rings were intact, and it was still covered with a tin lid. SW was found not dry before rainwater infiltration started, with an initial water column of about 0.7 m. During preliminary cleaning, the mud was removed from the bottom before the start of the experiment and once again in May 2021 to maintain the infiltration efficiency of SW. Since the rainwater was introduced into SW solely driven by gravitation in the experiment (i.e., no pumps were used), the term *"infiltration"* describes better the rainwater harvesting process rather than *"injection"*.

The overall inclined rooftop area was 115 m^2; considering vertical rainfall, this corresponded to about 80 m^2 of projected flat surface receiving rainfall. Thus, the potentially collectable amount of vertical rainfall was 80 m^2 multiplied by the precipitation rates.

The shallow dug well was connected to the rooftop gutters by PVC hoses. The gutters were cleaned twice during the experimental time: before the experiment started and in December 2020. The water passed through a filter mesh before it entered the tube system to impede the entrance of leaves and other larger objects into the well. The filter mesh did not, however, remove fine grained materials. The filters were changed in December 2020 and May 2021.

Two monitoring wells, P1 and P2, were constructed in the direction of groundwater flow for water level observation and for sampling purposes, as schematically shown in Fig. 2. The radial distance between SW and P1 is 8.3 m and between SW and P2 is 12.3 m. The depth of the monitoring wells is 6.7 m, of which the last 1 m is screened. The diameter of the wells is 0.04 m.

3. Material and methods

Understanding the shortcomings of previous field experiments regarding rooftop rainwater harvesting coupled with shallow wells; water level, temperature and specific electrical conductivity were recorded continuously every half hour in SW and in the monitoring wells by automatic data loggers to collect information about the changes in these parameters, and thus the impact of the rainfall harvesting process on native groundwater. In addition, meteorological data (precipitation and daily average temperature) were available from Időkép Kft., measured in Kerekegyháza (between January 28, 2020 and June 29, 2021) and from the Hungarian Meteorological Service (OMSZ), measured in the neighboring village, Fülöpháza (from June 30, 2021). Complimentary water level monitoring data of 5 shallow monitoring wells were used which were acquired from the territorially competent water directorate (ADUVÍZIG) to compare them with the time-series data measured in SW, P1 and P2 during the experiment. The

Table 1
Summary of measured parameters, the place of measurements, used methods and their accuracy.

Laboratory/place of measurement	Parameter	Measuring interval	Method	Accuracy/detection limit
On site (SW)	Water level EC on 25 °C Temperature	31/01/20–21/09/21 (30 min)	DATAQUA measuring device	±0,1% ±1% ±0,1 °C
On site (P1, P2)	Pressure EC on 25 °C Temperature	04/02/20–21/09/21 (30 min)	CTD-Diver measuring devices	±0.5 cmH_2O ±1% ±0,1 °C
On site	Air pressure Air temperature	04/02/20–21/09/21 (30 min)	Baro DIVER	±0.5 cmH_2O ±0,1 °C
Department of Geology, ELTE Eötvös Loránd University, Budapest, Hungary	Ca^{2+} Mg^{2+} Na^+, K^+ HCO_3^- Cl^- SO_4^{2-} NO_3^-	SW: from 14/01/20 (12 times), P1, P2: from 10/09/20 (7 times), PR, RT: 14/04/21, KE01, KE02: from 21/09/20 (5 times)	EDTA titrimetric method EDTA titrimetric method Flame photometry Alkalinity titration Argentometric titrimetry Spectrophotometry Colorimetric test kit (VISOCOLOR ECO Nitrate)	±5 mg L^{-1} ±2 mg L^{-1} ±0.5 mg L^{-1} ±12 mg L^{-1} ±2 mg L^{-1} ±5 mg L^{-1} Gradation: 0-1-3-5-10-20-30-50-70-90-120 mg L^{-1}
Institute of Aquatic Ecology, Centre for Ecological Research, Budapest, Hungary	TOC, DOC	SW, P1, P2: 11/02/21, PR, RT: 14/04/21	MULTI N/C 3100 TOC/TN analyser (Analytik Jena, Jena, Germany)	±3%
Mining and Geological Survey of Hungary, Budapest, Hungary	Ca^{2+}, Mg^{2+}, Na^+, K^+, Mn^{2+}, Fe^{2+}, PO_4^{3-} HCO_3^-, CO_3^{2-} Cl^-, SO_4^{2-} NO_3^-, NO_2^- NH_4^+ F^- H_2SiO_3 Trace elements COD TSS	SW, P1, P2: from 11/02/21 (5 times), PR, RT: 14/04/21, KE01, KE02: 21/09/21	Jobin-Yvon Ultima 2C ICP-OES Acidimetry (calculated) Ion chromatography – CD Ion chromatography – UV Spectrophotometry Photometry Jobin-Yvon Ultima 2C ICP-OES Elmer ELAN DRC II ICP-MS Permanganometry Weight measurement	Detailed information available from the laboratory ±0.5 mg L^{-1} ±2 mg L^{-1}
Hertelendi Laboratory of Environmental Studies (HEKAL), Institute for Nuclear Research – Isotoptech Ltd., Debrecen, Hungary	δ^2H $\delta^{18}O$ ^3H	SW, P1, P2: from 15/12/20 (5 times) SW, P1, P2: from 15/12/20 (3 times)	Cavity Enhanced Laser Spectroscopy ^3He ingrowth method	±0.50‰ ±0.08‰ ±0.15 TU

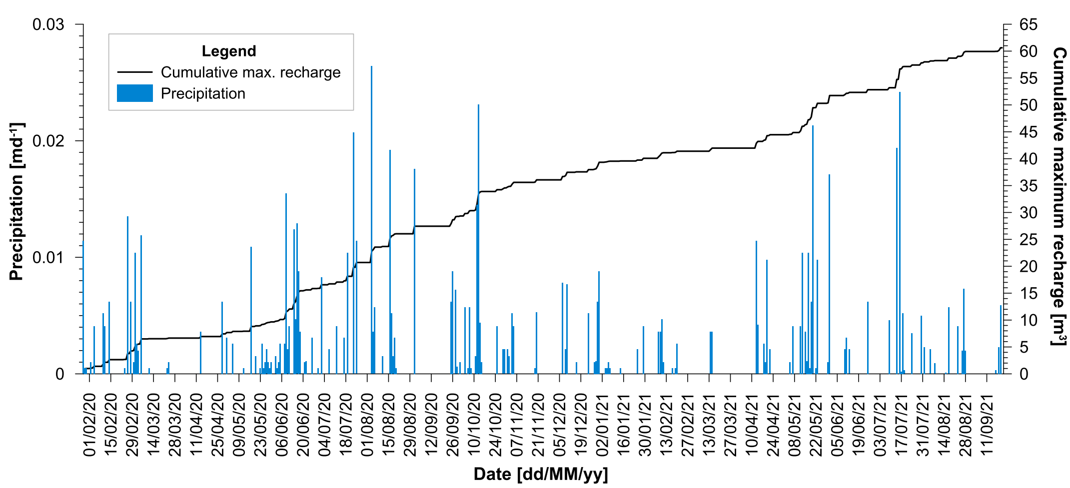

Fig. 3. Daily precipitation rate [md^{-1}] and cumulative maximum recharge [m^3] during the monitoring period (data source: Időkép – until June 29, 2021, OMSZ – from June 30, 2021).

maximum distance of these wells from the pilot site was 5 km (Fig. 1c).

In order to get to know the shallow unconfined environment, sediment samples were collected from the aquifer during drilling of P1 (sample "DR-A") and from the bottom of SW (samples "SW-T″ from the upper layer and "ST-B″ from the lower layer) during maintenance (May 2021). Grain-size measurements were carried out by sieving (above d = 0.063 mm) and elutriation (below d = 0.063 mm) in the laboratory of Department of Geology, ELTE Eötvös Loránd University (Budapest, Hungary). The organic matter can influence contamination fate and transport; therefore, it was estimated for SW-T and ST-B by measuring loss on ignition (LOI) at 550 °C. Hydraulic conductivity (K) values were determined using the Excel-based tool called HydrogeoSieveXL (Devlin, 2015). Hydraulic conductivity of the well bottom was also estimated by evaluating infiltration curves, for 22 selected precipitation events, as slug tests using the AquiferTest 11.0 Pro software (WH, 2021). The Bouwer & Rice method (Bouwer and Rice, 1976) was found to be the most applicable for the experiment. The following parameters were used for the evaluation: unconfined aquifer, aquifer thickness: 20 m, partially

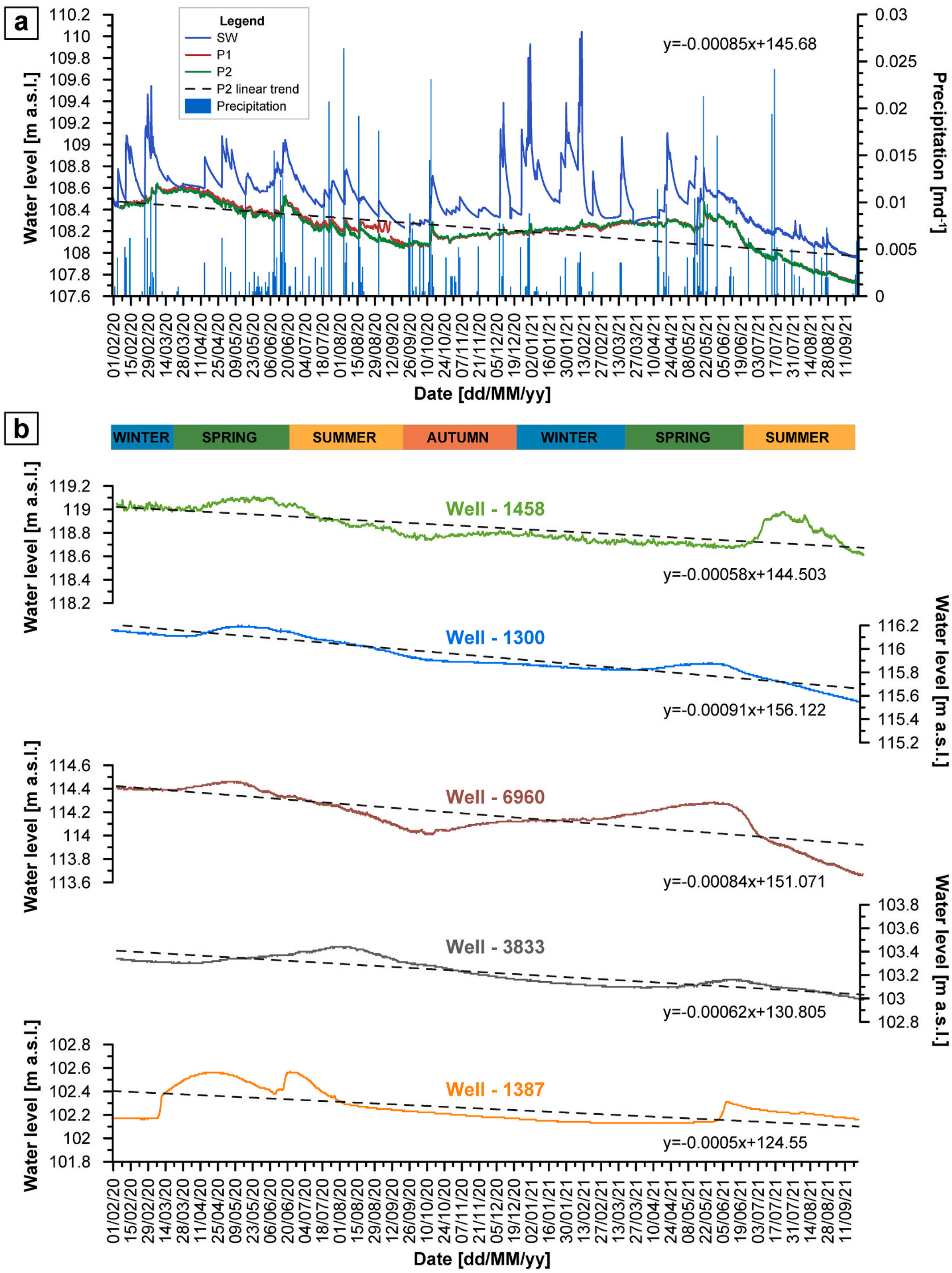

Fig. 4. Water level changes in (a) the shallow well (SW) and the monitoring wells (P1, P2) of the experiment and in (b) the observation wells of the broader area (data source: ADUVÍZIG). Linear trends fitted to P2 (Fig. 4a) and to the observation wells (Fig. 4b) are plotted with dashed lines and the equation of the lines are also given. Daily precipitation is indicated on Fig. 4a (data source: Időkép – until June 29, 2021, OMSZ – from June 30, 2021). On Fig. 4b astronomical seasons are indicated.

penetrating well, well diameter: 0.4 m, screen length: 0.2 m, well depth: 6.2 m, porosity: 30%.

Water sampling was conducted seasonally by a submersible pump in the case of SW, after taking out approx. 300 L of water. P1 and P2 were sampled with small bailers (volume: 0.1 L), due to their small diameters, after taking out 1 L of water. Water samples were collected from SW from January 2020, and from P1–P2 from September 2020 for measuring the main anions and cations in the laboratory of Department of Geology, ELTE Eötvös Loránd University (Budapest, Hungary).

Following the interaction of rooftop rainwater with shallow groundwater, stable isotope ($\delta^{18}O$ and $\delta^{2}H$) and tritium measurements started in December 2020 at Hertelendi Laboratory of Environmental Studies (HEKAL) (Debrecen, Hungary). In addition, detailed laboratory analysis of water samples, including main and trace elements, chemical oxygen demand (COD) and total suspended solids (TSS) content as well, started in February 2021 and continued seasonally at the Mining and Geological Survey of Hungary (Budapest, Hungary). Precipitation was sampled once in April 2021. Two samples were taken: PR – precipitation

Table 2
The observed water level difference [m] in the wells taking part in the experiment (SW, P1, P2) and other observation wells in the area.

Well ID	Water level [m a.s.l.] difference in September			Difference in yearly maximum water levels [m a.s.l.]		
	September 21, 2020	September 21, 2021	Difference	Max 2020	Max 2021	Difference
SW	108.24	107.99	−0.25	–	–	–
P1	108.18	107.84	−0.35	108.74	108.53	−0.21
P2	108.15	107.83	−0.32	108.74	108.57	−0.17
W-1458	118.77	118.61	−0.16	119.11	118.98	−0.13
W-1300	115.94	115.55	−0.39	116.2	115.88	−0.32
W-6960	114.04	113.66	−0.38	114.46	114.29	−0.17
W-3833	103.3	102.99	−0.31	103.44	103.16	−0.28
W-1389	102.23	102.16	−0.07	102.57	102.31	−0.26
Average	–	–	−0.28	–	–	−0.22

collected directly in buckets, RT – rooftop rainwater collected from the PVC hoses. Total organic carbon (TOC) and dissolved organic carbon (DOC) content of SW, P1, P2, PR and RT were measured once at the Institute of Aquatic Ecology, Centre for Ecological Research (Budapest, Hungary).

Considering the aims of the research, these measurements were used i) to determine the original water composition, ii) to monitor the occurring changes during the experimental period, iii) to follow the infiltration process by comparing the data sets of SW, P1 and P2, iv) to compare the results with the Hungarian limit values for groundwater (Decree, 2009) in order to delineate possible contaminants.

Two control wells (KE01, KE02; Fig. 1c) were involved in the evaluation, where rainwater is directed to the well by the residents for ~30 and ~10 years respectively. They were sampled from September 2020 seasonally for main elements and once for detailed laboratory analysis in September 2021 (main and trace elements as well) to identify any long-term contamination risk. The water was sampled with a submersible pump, after taking out approx. 300 L of water, similarly to SW.

Detailed summary of monitored parameters and related accuracies, the used methods and equipment can be found in Table 1.

4. Results and interpretation

4.1. Groundwater hydraulics and quantity

4.1.1. The recharge rate of SW

The daily precipitation rates (P) observed in the study area between January 2020 and September 2021 are shown in Fig. 3. Such P can be used to obtain an estimation of the collectable amount of rainwater that can artificially recharge the aquifer through SW. The volumetric recharge rate of SW can be calculated based on Equation (1):

$$Q_{SW} = A_r \bullet P \tag{1}$$

where Q_{SW} is the volumetric recharge rate of SW [m^3d^{-1}], A_r is the area of the rooftop [m^2] and P is the daily precipitation rate [md^{-1}]. Considering the area covered by the rooftop $A_r = 80$ m^2, an event with $P = 0.01$ md^{-1} would correspond to a volumetric recharge rate of SW of $Q_{SW} = 0.8$ m^3d^{-1}. This linear calculation can be used to compute the cumulative Q_{SW} occurring during the experimental time, which is shown in Fig. 3. The total amount of rainfall (P_{tot}) at the end of the monitoring period (a total of 20 months) was $P_{tot} = 0.7577$ m, thus resulting in $Q_{tot} = 60.616$ m^3 collected rainwater that could be used to artificially recharge the aquifer. Note that the actual amount of rainwater falling on the rooftop and reaching the well is influenced by many factors, such as wind direction, rainfall intensity, rainfall length, evaporation rate and the conditions of the gutter and PVC hose systems. However, we consider that the obtained values are a valid first-cut indication of the scale of rainfall volumes that were recharging the aquifer through SW.

4.1.2. Temporal changes of water level

The water level in SW, P1 and P2 showed seasonal changes (Fig. 4a), with an increase until the end of March 2020, a decrease until mid-September 2020, then an increase until mid-May 2021 and a decrease until the end of the experimental time (end of September 2021). The aquifer response in SW is directly linked to the occurrence of the rainfall events (i.e., rainfall-driven aquifer recharge). Water level in SW is always higher than in P1 and P2, as it is the infiltration well and located slightly upgradient (Fig. 2). The water level maximum of P1 and P2 occurs in March in 2020 and in May in 2021. The range of water level change in SW is approx. 2 m, while that in P1 and P2, is approx. 1 m.

The water level time series of SW, P1 and P2 were compared with the water level time series of shallow observation wells in the vicinity of Kerekegyháza (for location see Fig. 1c) to evaluate the similarities and differences in the observed trends. An overall water level decrease can be observed in all of the wells (Fig. 4 and Table 2). On average, water levels decreased 0.28 m in one year (between 21 Sept. 2020 and 2021). The rate of decline was 0.25 m, 0.35 m and 0.32 m over one year for SW, P1 and P2 respectively. The least effected well is W-1387 (located at the local discharge area), where water level decreased only 0.07 m in a year. The water level maximums decreased as well, on average 0.22 m. While in the experimental wells, the rate of decrease was lower, 0.21 m and 0.17 m for P1 and P2, respectively.

Considering the temporal differences in the water level maximum of the wells, in 2020, the water level maximum occurs much sooner in SW, P1, and P2, than in the other wells (May: W-1458, W-1300, W-6960, July–August: W-3833), except for W-1387. In 2021, the water level maximums are observed in May–June, except for W-1458 which shows the highest values in July. The autumn-winter water level increase seen in the case of SW, P1 and P2, does not or only less markedly occurs (with the exception of W-6960). The behavior of the wells taking part in the experiment is very similar to the other observation wells in the area, however the discovered differences, namely the earlier water level maximum and the autumn water level increase, might be caused by the rooftop rainwater infiltration.

4.1.3. Changes in water column increase

To better understand the dynamics of rainwater infiltration through SW, the induced water columns were evaluated. The water level in SW is very sensitive to the precipitation events (Fig. 4a). Similarly to a slug test, the water level quickly increases when rainfall events occur, as a consequence of the direct funneling of rooftop-collected water inside the well. After the precipitation events, the water level decreases proportionally to the hydrodynamic properties of the aquifer, specifically the hydraulic conductivity (K) and the specific yield (S_y).

The water level of SW shows a sudden increase and then a gradual decrease (recession) due to a precipitation event as the water seeps into the aquifer (Fig. 4a). However, the precipitation rate (P) was not always linearly proportional to the increase of the water column (ΔH) induced by the funneled rooftop-collected water. For analyzing the relationship of P and ΔH, 58 distinct precipitation events were selected, each of them lasting for maximum one day (Fig. 5).

Comparing P and ΔH, two different correlations can be observed (Fig. 5a). The different seasons are showing different relationships: in

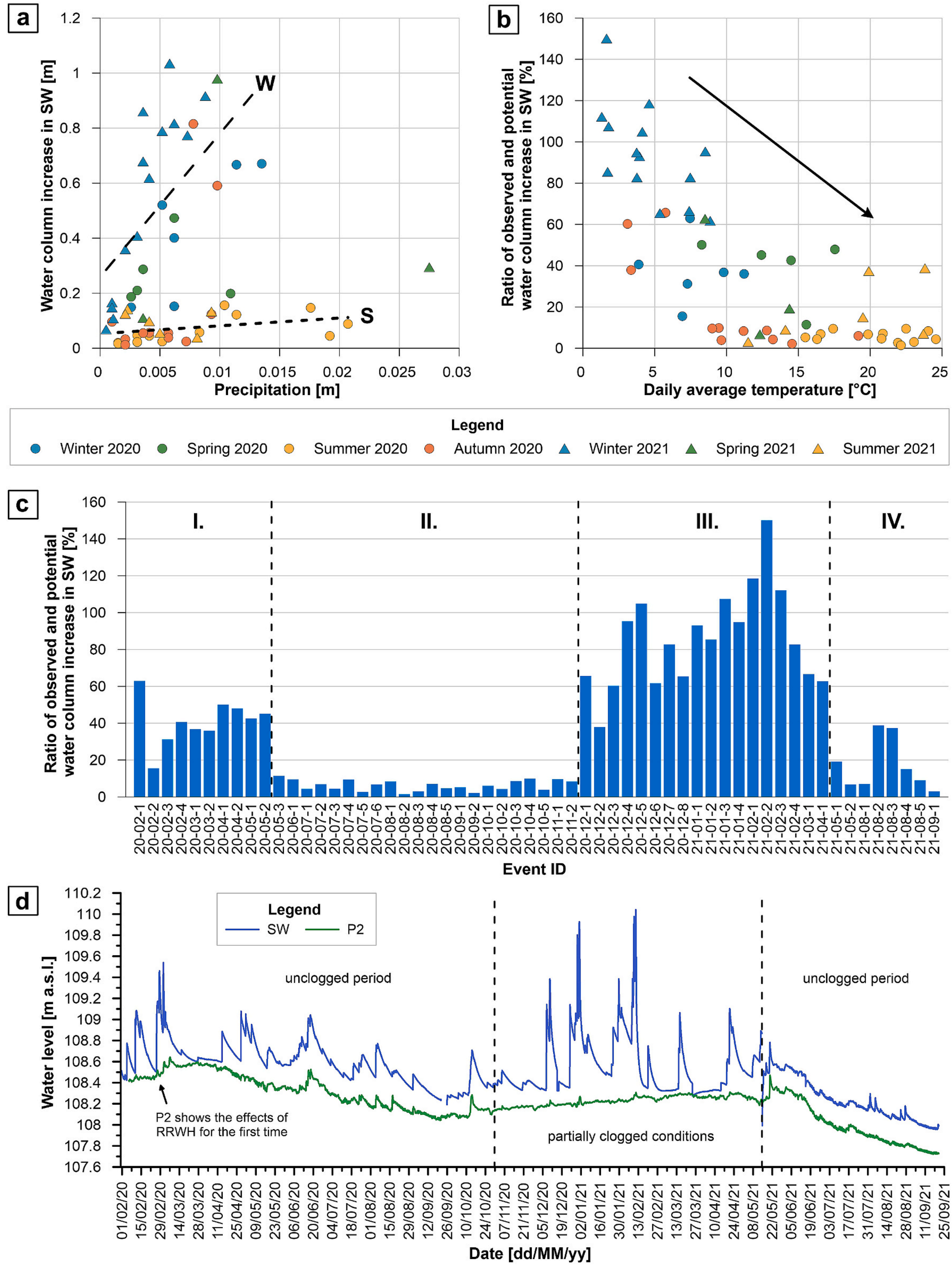

Fig. 5. (a) The relationship between precipitation rate (*P*) and water column increase (*ΔH*) in SW with linear regression lines fitted to winter samples (W) and summer samples (S). (b) The relationship between daily average temperature [°C] and the ratio of observed and potential water column increase [%] in SW. The precipitation events are categorized based on astrological seasons of different years. (c) Changes over time in the ratio of observed and potential water column increase [%] in the shallow well (SW) for selected precipitation events. Event ID: yy-MM-number. (For calculations see Chapter 4.1.3.). (d) Water level changes in SW and P2 complemented by the different clogging rates.

winter the water column increase is higher, while in summer it is lower (Fig. 5a), spring and autumn events are falling into both of the groups.

If the potentially collectable amount of rooftop water fully reaches the well, theoretically, water level increases proportionally. Based on this, the following equation (Equation (2)) was introduced for the calculation of potential water column increase:

Table 3
Hydraulic conductivity of the aquifer (DR-A), based on the sample collected during drilling of P1 and the sediments collected from the well bottom of SW (SW-T: upper layer, SW-B: lower layer).

Sample ID	Method				Mean	
	Sauerbrei [m s^{-1}]	Zunker [m s^{-1}]	Barr [m s^{-1}]	Alyamani and Sen [m s^{-1}]	Geometric mean [m s^{-1}]	Arithmetic mean [m s^{-1}]
DR-A	3.5E-06	1.2E-05	1.7E-06	1.6E-07	1.8E-06	4.4E-06
SW-T	1.3E-06	4.9E-06	1.7E-07	5.5E-07	8.7E-07	1.7E-06
SW-B	3.3E-07	2.2E-06	3.7E-08	8.5E-07	3.9E-07	8.5E-07

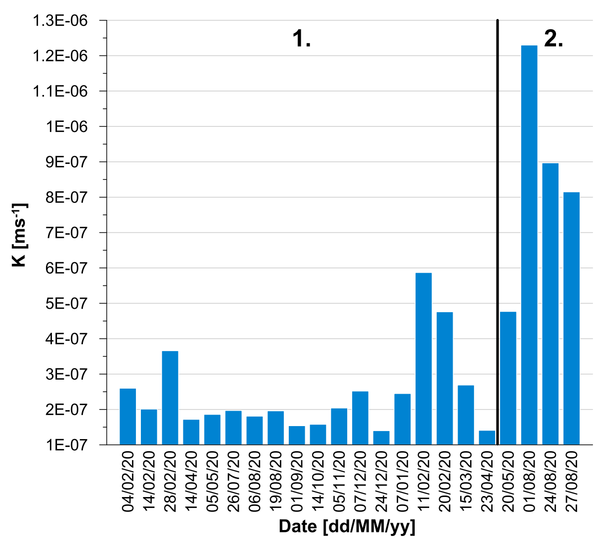

Fig. 6. Hydraulic conductivity (*K*) values based on the evaluation of 22 selected infiltration curves with Bouwer and Rice method (Bouwer and Rice, 1976). (1: before sediment removal from the well, 2: after sediment removal from the well).

$$\Delta H_{pot} = \frac{P \bullet A_r}{r_{SW}^2 \bullet \pi} \quad (2)$$

where ΔH_{pot} is the potential water column increase [m], P is the daily precipitation rate [md^{-1}], A_r is the rooftop area [m^2] and r_{SW} is the radius of SW [m]. As an example, theoretically 0.01 m precipitation induces 1.6 m water column increase. However, this exact relationship is rarely observed.

By comparing the ratio of the observed (ΔH) and the potential (ΔH_{pot}) water column increase for the selected events with the daily average temperature, a significant negative correlation (r = −0.74, p-value<0.05) was obtained (Fig. 5b). Higher temperatures can result in higher evaporation rates, especially during summer, thus less water can get into the well from the rooftop than in winter. This can be one influencing factor, however, evaporation solely does not explain the observed changes, as smaller ratios of ΔH and ΔH_{pot} can be observed even in October and November, when evaporation is not high (Fig. 5c).

By evaluating the temporal distribution of the ratio of ΔH and ΔH_{pot} for the selected events, the following trends can be noticed (Fig. 5c): Until May 2020, the average ratio of these values is 41% (Period I.), it's 6% during summer and autumn until December (Period II.), 86% until May 2021 (Period III.) and then decreased again to the average of 17% until September 2021 (Period IV.). Note for the interpretation, that the calculations are less reliable from Event 21-08-01, as the precipitation data were measured at a different meteorological station (see Chapter 3 for details). Additionally, the amount of water reaching the well is not measured, but estimated and dependent on many factors (see Chapter 4.1.1 for details), thus in some cases, the ratio of ΔH and ΔH_{pot} can be higher than 100%.

Assuming that the system is working properly and all the rainwater from the rooftop enters the well, the degree of water column rise depends on the rate of infiltration from the well to the aquifer during the precipitation event. Based on Darcy's Law, the infiltration rate depends on the hydraulic head difference (thus on the amount of precipitation in the well and the location of the water level in its surroundings) and on the saturated hydraulic conductivity (*K*) of the aquifer (Freeze and Cherry, 1979).

The decrease of the ratio of observed and the potential water column increase occurs in May in both years (Fig. 5c) after relatively low precipitation amounts in March and April (see Fig. 4a). These drier periods could result in lower water tables and drier conditions in the aquifer itself as well, which could have an impact on the infiltration rates. When the water table is lower and the pores are less saturated, the infiltration from the shallow well can be more effective, thus the water column increase in the well is lower. This process, however, cannot be well detected with the selected 30-min measuring frequency.

The influence of hydraulic conductivity can be best observed on Fig. 5c. There is a notable difference between Period I. and III. After 15.5 months of operation, a 40 cm thick layer of fine sediments accumulated in the bottom of the well, possibly decreasing the hydraulic conductivity due to clogging effect. Therefore, in Period III., the infiltration rate was slower, so a larger water column could form (Fig. 5c).

P1 and P2 also showed a response to precipitation events but to a lesser extent (Figs. 4a–5d). There were precipitation events when the water level increase in the monitoring wells was unambiguous (e.g., summer of 2020) and there were events when natural (daily) water level fluctuations masked the effect of rainwater infiltration.

Clogging process could also be detected by comparing SW and P2 levels (Fig. 5d). (There's no significant difference between P1 and P2 water levels, thus P2 was chosen for demonstration, as this well provided more reliable results.) The first observable increase in P2 water level occurred on February 26, 2020, one month after the project started. Between May and October 2020, the water level in P2 followed the water level of SW with higher peaks. Between October 2020 and May 2021 (time of well maintenance), SW showed much higher peaks, however, the water level of P2 barely increased after the precipitation events. It can be assumed that clogging process advanced in SW during autumn 2020 and slowed down the infiltration process and impaired the connection between the wells. However, the well bottom did not clog entirely, infiltration still occurred, since the water level of SW declined after every precipitation event. After cleaning the well bottom of SW, the water levels of the wells changed similarly again, without high peaks. These results well correspond to the ones obtained from the comparison of observed and potential water column increases in SW (Fig. 5c).

4.1.4. Infiltration curve analysis and changes in hydraulic conductivity

One of the key parameters that determines the efficiency of shallow well infiltration is the hydraulic conductivity of the well bottom. Based on grain-size distribution analysis, the hydraulic conductivity of the aquifer on average (geometric mean) is $1.8 \bullet 10^{-6}$ m s^{-1}, while the well bottom sediments (after 15.5 months of infiltration) showed $8.7 \bullet 10^{-7}$ m s^{-1} in the upper part and $3.9 \bullet 10^{-7}$ m s^{-1} in the lower part (Table 3). The hydraulic conductivity of the well bottom was also estimated by infiltration curve analysis (slug test) with Bouwer and Rice method (Bouwer and Rice, 1976). The result of each evaluated infiltration curve is plotted in chronological order in Fig. 6 to follow the temporal changes in hydraulic conductivity, thus the effectiveness of infiltration. Before

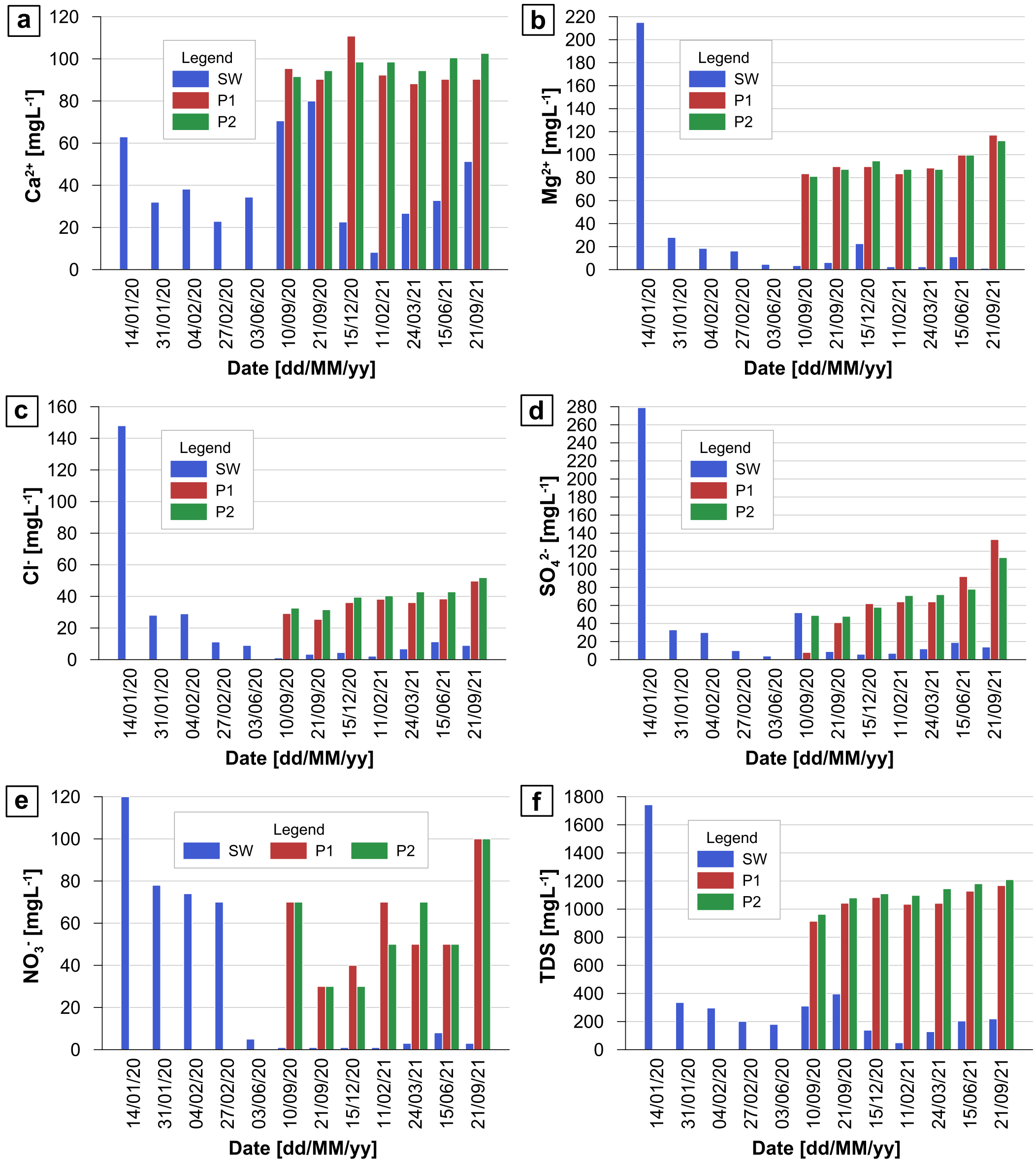

Fig. 7. The changes in the concentration of (a) Ca^{2+}, (b) Mg^{2+}, (c) Cl^-, (d) SO_4^{2-}, (e) NO_3^-, and in (f) total dissolved solids content (TDS) in the shallow well (SW) and in the monitoring well (P1, P2). Note: The limit of quantification for sulfate is 25 mg L^{-1}.

cleaning the well bottom (May 2021), most of the events showed *K* values between 1 and $3{\bullet}10^{-7}$ m s^{-1}, with the exception of 3 winter precipitation events. After the accumulated sediments were removed, the hydraulic conductivity increased, even reaching $1.2{\bullet}10^{-6}$ m s^{-1} (Fig. 6). These results are in accordance with the ones obtained from grain-size distribution analysis and from the evaluation of water column changes (Chapter 4.1.3).

4.2. Consequences on groundwater quality and temperature

4.2.1. Composition of main ions in water

First of all, baseline conditions were surveyed by sampling SW on January 14, 2020, before the project started (Fig. 7, Table A1). The shallow water composition showed Mg^{2+} and HCO_3^- dominance. The total dissolved solids content (TDS) was considerably high (1743 mg L^{-1}), as well as Mg^{2+}, Na^+, Cl^-, SO_4^2, NO_3^- concentrations. SW displayed a significant improvement in water quality during the experiment (Fig. 7, Table A1-2): Mg^{2+}, Na^+, Cl^-, SO_4^{2-}, NO_3^- concentrations and TDS decreased significantly due to rainwater infiltration. An initial decrease of 87%, 82%, 81%, 88%, 35% and 81% (respectively) was observed after the infiltration started (sampling time: 31/01/20) and a further 96%, 93%, 68%, 58%, 96% and 35% decrease occurred in these parameters by the end of the monitoring period (sampling time: 21/09/21). Initially, NO_3^- exceeded the limit value for shallow groundwater (50 μg L^{-1}, Decree, 2009), but from June 2020, the values complied with the legislation. While Ca^{2+} and HCO_3^- concentrations (and TDS to some extent) were constantly changing in time, depending on the rainfall amount reaching the well.

The first measurements of P1 and P2 occurred in September 2020, 7 months after the project started (Fig. 7, Table A1). In contrast with the decreasing trends observed in SW, a slight increase in Mg^{2+}, Na^+, Cl^-, SO_4^{2-}, NO_3^- concentrations and increasing TDS were detected. NO_3^- displayed the highest variability in this regard.

4.2.2. Trace elements and possible contaminants

The quality of precipitation (PR) and rooftop water (RT) were examined, and the results mostly complied with the Hungarian legislation regarding groundwater (Table A2; Decree 6/2009), except for NH_4^+

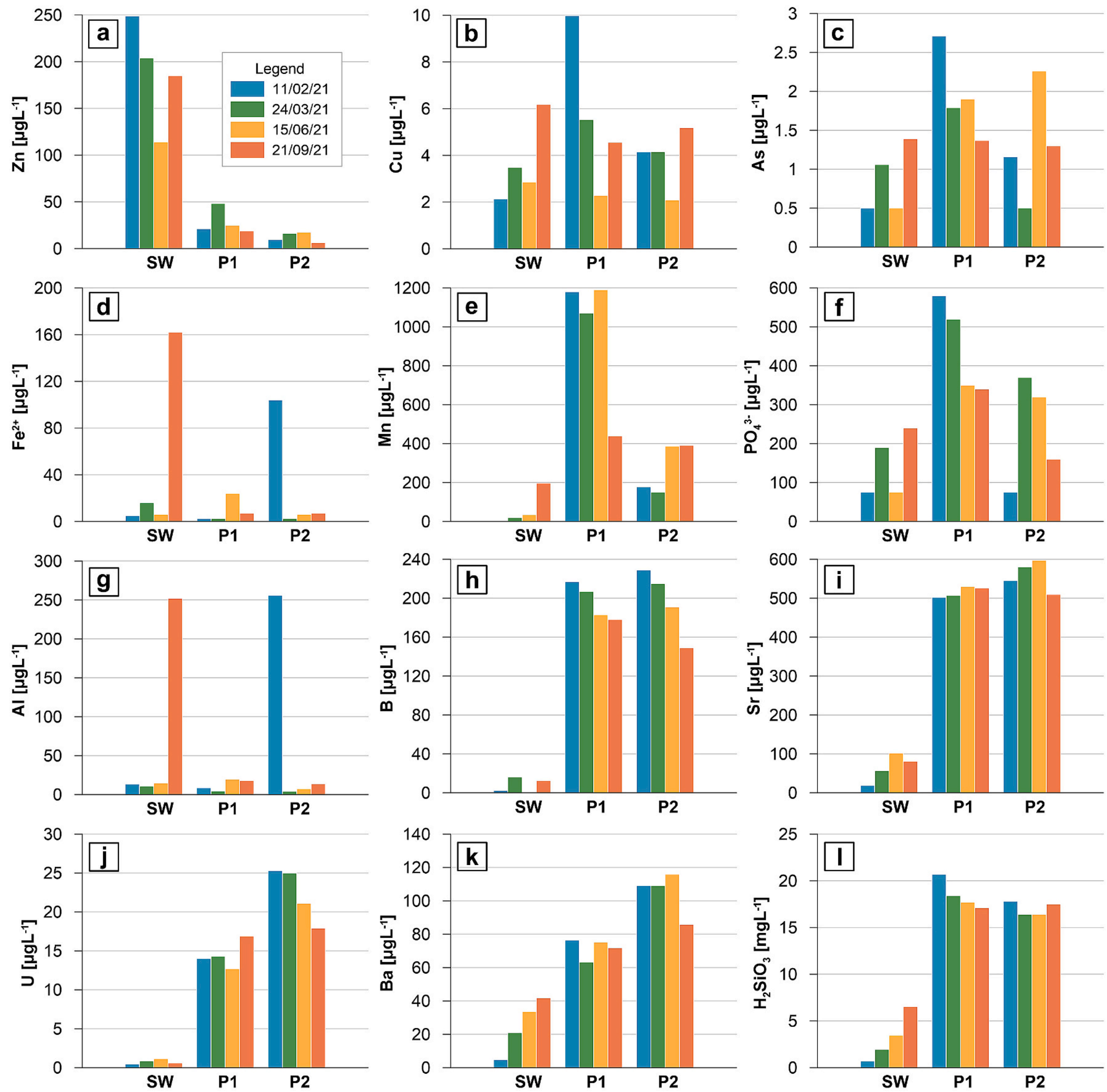

Fig. 8. The changes in the concentration of selected trace elements and components in the shallow well (SW) and in the monitoring wells (P1–P2). (a) Zinc, (b) copper, (c) arsenic, (d) ferrous iron, (e) manganese, (f) phosphate, (g) aluminum, (h) boron, (i) strontium, (j) uranium, (k) barium and (l) silicic acid (in mg L^{-1}).

and Zn. NH_4^+ was slightly higher in PR, than the limit value (0.5 mg L^{-1}), however, higher concentrations were not detected in SW in any of the samples. In RT, only zinc concentration exceeded the limit value (200 μg L^{-1}), which was also notably high (114–249 μg L^{-1}) in SW (Fig. 8a), but overall, it decreased during the observation period. Based on the comparison of PR and RT samples, rooftop water was enriched in a number of trace elements compared to rainwater (Table A2). Zinc concentration was 48.2 times higher in rooftop water. In case of Sr, Cu, Mn, Ba and Al, it was about 6.9, 6, 4.3, 3.6 and 1.9 times higher in roof runoff, respectively. These results indicate that, the roof material, the gutters and the tube system are affecting the water quality.

Furthermore, DOC, TOC, COD and TDS are all higher in the rooftop water than in the precipitation. On the other hand, in the shallow well, the values are not elevated, so the sedimentation (and/or degradation) of organic matter on the bottom of the well can be assumed. The sedimentation process was also confirmed when the well bottom was cleaned in May 2021. A 40 cm thick mud layer accumulated on the bottom after only 15.5 months of infiltration. The organic content of the samples was not significant, LOI was 3.41% for the upper layer (SW-T) and 2.53% for the lower layer (SW–B). In the monitoring wells, TOC and TSS content is high compared to PR, RT and SW. It might have a natural (geological) reason, or it might be due to the design of the wells. COD values are similar in SW and in P1–P2.

In SW, water quality complied with the Hungarian limit values for groundwater with zinc being the only exception (Table A2; Decree 6/2009). In P1 and P2, NO_3^- concentrations were significant (81–165 mg L^{-1}) and in some cases NH_4^+ and PO_4^{3-} exceeded the limit.

Comparing the trace element composition of SW and P1–P2, the following trends can be observed: Zinc concentrations are significantly higher compared to P1 and P2. While it decreased in SW, it fluctuated in the monitoring wells (Fig. 8a). Copper and arsenic concentrations were constantly changing in all three wells, not showing an exact trend (Fig. 8b and c). They slightly increased in SW and decreased in P1 however, their concentrations were well below the limit values (200 μg L^{-1} for Cu and 10 μg L^{-1} for As). Manganese concentrations were quite low in SW but showed an increase. The values were one order of magnitude higher in P1 and decreased by the end of the monitoring period. P2 values were falling in between and also increased (Fig. 8e). In contrast, ferrous iron and aluminum concentrations were low in all three wells, except for 1-1 occasional increase in SW and P2 (Fig. 8d,g). Phosphate showed an increase in SW, while it decreased in P1 and in P2, after the second sampling (Fig. 8f). Boron showed very low

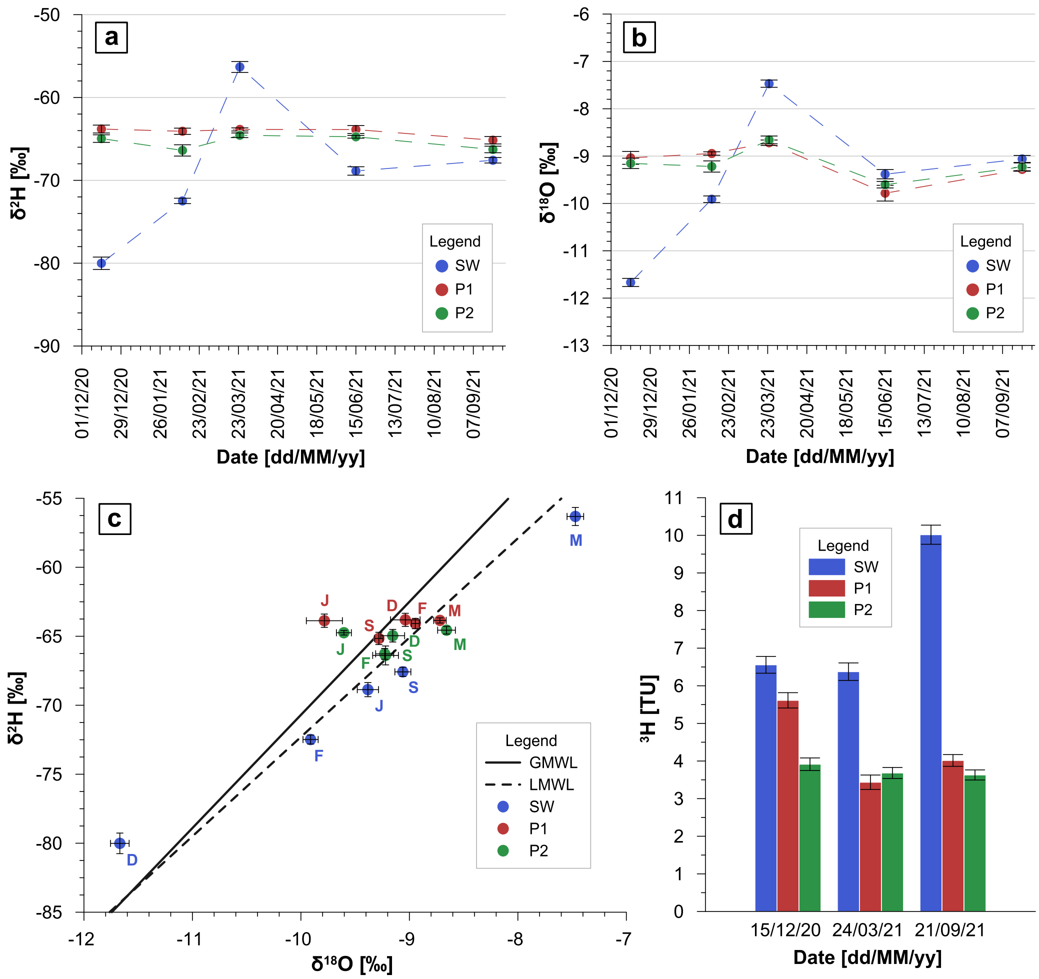

Fig. 9. Changes in (a) δ^2H, (b) δ^{18}O, (c) δ^2H–δ^{18}O and (d) ^{3}H in the shallow well (SW) and in the monitoring wells (P1–P2). The letters on Fig. 9c indicate the sampling times: D–15/12/20, F–11/02/2021, M–24/03/2021, J–15/06/21, S–21/09/21. Global Meteoric Water Line (GMWL): $\delta^2H = 8.2\delta^{18}O + 11.3$ from Rozanski et al. (1993), Local Meteoric Water Line (LMWL): $\delta^2H = 7.2\delta^{18}O + 0.3$ from Bottyán et al. (2017).

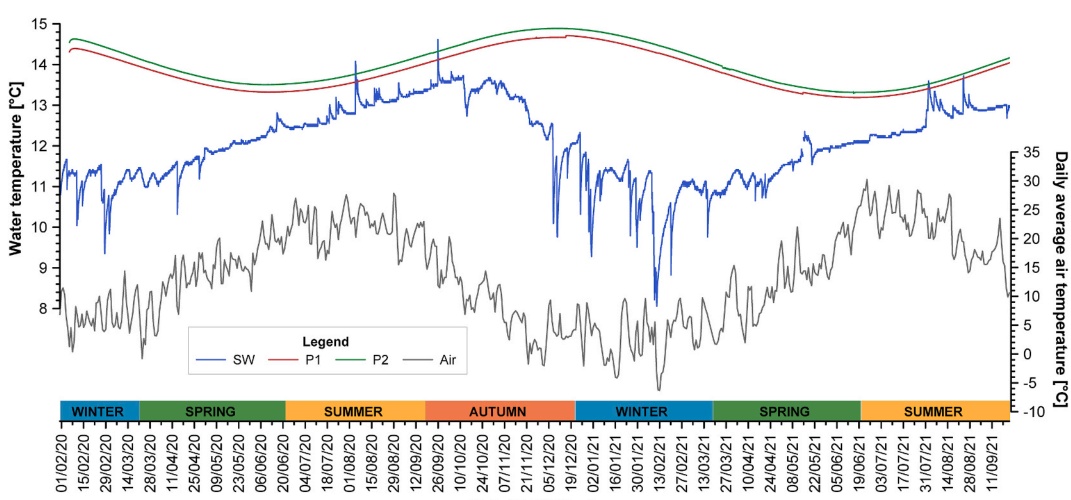

Fig. 10. Temperature changes in the shallow well (SW), the monitoring wells (P1, P2) and changes of daily average air temperature (data source: Időkép – until June 29, 2021, OMSZ – from June 30, 2021). On the bottom of the figure, astronomical seasons are indicated.

concentrations in SW and significantly higher ones in P1 and P2, additionally, a constant decrease can be observed in the latter ones (Fig. 8h). Uranium showed a similar picture, but a decrease can only be observed in P2 (Fig. 8j). Silicic acid content was also lower in SW and increased during the monitoring period, while it was higher in P1–P2 and showed a decrease only in P1 (Fig. 8l). Although barium concentrations increased as well in SW, there was no observable trend for P1 and P2 (Fig. 8k). Strontium concentrations changed similarly in all three wells during the monitoring period, however, SW showed significantly lower values than P1 and P2 (Fig. 8i).

4.2.3. Water chemistry of the control wells

We used the data of two additional shallow wells (control wells) with rainwater infiltration from the surroundings of the experimental area (Fig. 1c) for comparison to better understand the long-term effect of rainwater infiltration on groundwater quality. The water composition of KE02 was quite stable while that of KE01 showed seasonal variations. The control wells were showing very similar compositions to SW (Table A1): low TDS and low Mg^{2+}, Na^{+}, Cl^{-}, SO_4^{2-}, NO_3^{-} concentrations. On the other hand, in both wells the PO_4^{3-} concentration exceeded the limit value for groundwater (0.5 mg L^{-1}). All other values were compliant with the legislation. The zinc values (38 and 70 μg L^{-1}) were lower than in SW, but higher than in P1 and P2. COD was also slightly higher in the control wells than in SW, P1 and P2. This can be the cause of sediment load from the roof to the wells because there are no filters built-in these systems.

4.2.4. Isotopic changes

Isotope measurements were included in the research to follow the effects of the infiltration process. The observed $\delta^{2}H$ and $\delta^{18}O$ trends in the wells showed similar patterns, especially in case of $\delta^{18}O$ (Fig. 9a and b). The range of change was much higher in SW, while it was lower for P1 and P2. Similar changes can be observed on Fig. 9c as well. This shows the effect of rooftop water inflow to the infiltration well. Significant effect of evaporation cannot be seen. For SW, the lowest values can be observed in December 2020 and the highest ones in March 2021, both in terms of $\delta^{2}H$ and $\delta^{18}O$. For P1 and P2, the values of $\delta^{2}H$ were barely changing, while $\delta^{18}O$ showed seasonal changes. The lowest values were observed in June 2021 and the highest ones in March 2021. The measured values are close to the global (GMWL) and local meteoric waterlines (LMWL), with the exception of the samples taken from SW in December 2020 and from P1 and P2 in June 2021 (Fig. 9c).

Tritium increased in SW, decreased in P1 and was quite stable in P2 (Fig. 9d). At the depth of P1 and P2, tritium values of 3.5–4.0 TU are expected in the study area (Palcsu et al., 2017). The elevated tritium concentrations in SW can be attributed to the contribution of the precipitation, which has an annual variation between 5 and 15 TU (Palcsu et al., 2018). The highest tritium values of 10 TU sampled in September 2021 can be explained by the inflow of summer precipitation with higher tritium concentration.

4.2.5. Water temperature changes

Water temperature changes of the wells were compared with each other and with the daily average air temperatures to evaluate the effect of rooftop rainwater infiltration. SW showed lower temperature values than P1 and P2 (Fig. 10). The infiltrated water significantly affected the water temperatures in SW, which is represented by the positive (heating) and negative (cooling) anomalies in the dataset. The collected water had a cooling effect on well water until May 2020, then a heating effect until October 2020, a cooling effect until May 2021 and then a heating effect again until the end of the monitoring period (September 2021). On the other hand, seasonal changes are also observable in SW: water temperature increased until October 2020, decreased until February 2021 and increased again until September 2021. The range of temperature changes during the monitoring period is approx. 6.5 °C in SW and 1.5 °C in P1 and P2.

The air temperature reached its maximum in August, while the water temperature in the shallow well in October and that of the monitoring wells in December. The colder winter precipitation had a cooling effect on the water of SW, which can also indirectly cause cooling in P1–P2 wells by June. The water heats back up by December and then cools down again by June in these wells.

5. Discussion

Based on the interpretation of individual results and literature review, the most significant findings related to the field experiment can be discussed. The extension possibilities of the results were also evaluated for the area and for different regions worldwide.

5.1. Quantitative potential of RRWH-SW

All of the wells, including SW, P1, P2 and the observation wells in the vicinity of Kerekegyháza showed a decrease in water levels (Figs. 1c and 4), following the long-term trend of the area (i.e., declining water level; e.g., Garamhegyi et al., 2020). Although the water level in SW and in the monitoring wells (P1, P2) did not increase permanently during the monitoring period, the effect of infiltration has been confirmed. Comparing the water levels of SW–P1–P2 with other observation wells from the area showed that water level maximum occurs earlier in the wells which are part of the experiment, and the autumn-winter water level increase is only present in one other well apart from the experimental ones (Fig. 4).

Based on the results of analyzing the differences between the precipitation and water column increase ratios (Chapter 4.1.3), seasonality strongly affects the infiltration process. SW infiltrates the aquifer faster after dryer periods (i.e., drought) and when the water table is lower. If the summer precipitation can be collected in sufficient amount in the infiltration well, evaporation loss is reduced, and this water can effectively recharge the uppermost aquifer. Considering that the natural infiltration of precipitation in the Danube-Tisza Interfluve is only 14 ± 9% due to high evaporation rates (approx. 80–90%, Szilágyi et al., 2012), collecting rainwater and thus water retention in the saturated zone is highly important in the area.

Although, the infiltrated water amount during the observation period was only 60.616 m^3 (approx. 40 m^3y^{-1}), by involving more rooftops into the experiment, thus increasing the amount of harvestable rainwater, the impact of RRWH-SW on water levels would be more significant. Based on GIS-based settlement-scale calculations of Yousif (2022), the total roof-covered area in Kerekegyháza is ~0.55 km^2 (15% of the total area). Thus, calculating with an average of 515 mm annual precipitation, the total collectable water amount from the roofs of Kerekegyháza is 283,250 m^3y^{-1}, which is a remarkable potential. Based on simple water balance calculations, infiltrating all the collectable water from roofs would increase the groundwater table with approx. 0.27 m in one year, which is comparable with the yearly decrease of water table (Yousif, 2022). The collectable volume of rainwater can be further increased if surface runoff was collected from roads as well (Shekh, 2021). The treated wastewater of the settlement offers additional opportunities, however, for the safe reuse of these waters, further field experiments and water chemical measurements are needed in the future.

5.2. Qualitative potential of RRWH-SW

The observed concentration changes in SW, namely the decrease of Mg^{2+}, Na^{+}, Cl^{-}, SO_4^{2-}, NO_3^{-} and TDS, indicate significant improvement in shallow groundwater quality due to the infiltration of rainwater to the well (Fig. 7). Hasan et al. (2018) achieved similar results after 5 months of infiltration. While Dillon and Barry (2005) could only reach temporary water quality improvement in 2 years due to high TDS of native groundwater. In case of SW, these changes could be maintained, no significant increase was observable during the 20 months of the project. The measurements made in the control wells (KE01 and KE02) also confirmed that long-term improvement in water quality is achievable with this method. Thus, by infiltrating rainwater directly to the saturated zone, shallow groundwater can be diluted, therefore resulting in better quality. Rainwater stored underground can be abstracted later on for irrigation, if needed, as the water chemical parameters are adequate for that.

Groundwater quality improvement is not yet observable after 20 months in the monitoring wells (P1, P2) located 8.3 m and 12.3 m from SW. On the contrary, an increase in the concentration of these parameters can be seen. It can be assumed, that the surroundings of SW were contaminated before the experiment (Fig. 7, sampling time: 14/01/20) and the infiltrated rainwater pushed forward this water in the direction of local groundwater flow, thus in the direction of the monitoring wells. In the future, it is expected that these concentrations will start to decrease, similarly to the changes occurring in SW. Indicating this supposed process, a decrease in U, B, PO_4^{3-}, H_2SiO_3 concentrations occurs either in P1 or P2 or both (Fig. 8). These changes might be attributed to rainwater infiltration, but at this state of the research it cannot be stated with high certainty and further monitoring of water quality is needed.

Nevertheless, water samples from SW and the monitoring wells are well separated in terms of Zn, U, B, Sr and H_2SiO_3 concentrations (Fig. 8), so these parameters (and the main water composition) can be selected as "natural tracers" to monitor the movement of infiltrated water in the saturated zone. The effect of rainwater infiltration on stable isotope composition of the wells is already observable in SW (the range of change in δ^2H and δ^{18}O was higher in this well), however in P1 and P2 it is not yet evident (Fig. 9). Summer and autumn samples are very similar to each other, however, when more rainwater is accumulated (winter and spring), the wells are showing high differences in isotopic compositions. These differences might also be helpful in the future to track groundwater movement from SW, similarly to main and trace element compositions and groundwater temperature.

5.3. Environmental impacts of RRWH-SW

Sampling of precipitation (PR) and rooftop rainwater (RT) showed (Table A2) that despite enrichment in some trace elements (Zn, Sr, Cu, Mn, Ba and Al; Chapter 4.2.2), infiltrating rainwater through SW is able to dilute shallow groundwater and possible contamination is limited to only a few parameters.

Zinc not only occurs in rooftop rainwater in higher concentrations, but in SW as well. This is a common problem associated with rainwater harvesting systems, as the steel roofs or, in our case, gutters have an effect on the water composition (Meera and Mansoor Ahammed, 2006). Dillon and Barry (2005) had similar findings in Kingswood, Australia, where they observed 80–248 μg L^{-1} zinc levels in the rainwater storage tank, which they used for well infiltration. The control wells (KE01 and KE02), which are in operation for ~30 and ~10 years respectively, also showed elevated zinc levels (69.4 and 38 μg L^{-1}), however, they stayed well below the limit value (200 μg L^{-1}). These results indicate that the rainwater collecting systems are affecting the water quality of these wells too but considering that rainwater funneling to these wells takes place already for several years, zinc do not pose a high risk on groundwater quality in the long run. Contaminating groundwater with zinc is a possible risk related to RRWH-SW, thus it must be monitored in order to avoid any serious problems. Additionally, the decrease in Zn concentration in SW and lower concentrations measured in KE01 and KE02 can indicate the potential adsorption of Zn on clay minerals or bonding to organic matter at the bottom of the well, which has to be examined in the future in order to better understand the ongoing processes.

Several parameters showed concentration increase in SW, which can cause groundwater contamination, such as Cu, As, PO_4^{2-} and Ba, but their concentrations were well below the limit values, thus they do not pose a risk. Apart from the elevated Zn concentrations, the increase of Cu, Mn, Sr, Ba concentrations in SW can be attributed to rainwater infiltration, as rooftop-collected water showed enrichment in these elements (Table A2).

Ammonium only exceeded the limit value in the precipitation sample, but not in the samples taken from SW, thus it does not pose a considerable risk. KE01 and KE02 showed slightly elevated values of PO_4^{3-}, however, its source is not clear. It can either come from roof-runoff or can be the sign of previous contamination from leaking cesspools. Phosphorus and nitrogen are common component of rainwater harvested from roofs due to atmospheric deposition from transport, industrial processes and application of fertilizers (Novak et al., 2014).

5.4. Clogging

One of the reasons of why the authorities advise against RRWH-SW is clogging of the well bottom (Ministry of Interior, 2017). Clogging is a general process related to all MAR facilities (Dillon et al., 2016; Zhang et al., 2020) and it has to be managed, but it does not necessarily make the infiltration impossible. Clogging of the well bottom was identifiable through analyzing the water level time series of SW (Chapter 4.1.3 and 4.1.4) and during the maintenance of the well bottom. However, the infiltration during the experiment was successful, the accumulated mud at the bottom did not impede the infiltration significantly during the monitoring period. Although higher water columns could form in SW during partially clogged conditions, the collected water always seeped into the aquifer in relatively short time (max. 1–2 weeks depending on the precipitation rate). The reason behind is that higher water columns can even accelerate the infiltration rate by increasing the vertical hydraulic gradient, thus act against efficiency decrease induced by clogging. This is an advantage of using dug wells with large diameter and open bottom. The increased water columns in the infiltration well can be used for indicating the need for maintenance. If the ratio of ΔH and ΔH_{pot} is higher than 50–60% for 3 consecutive precipitation events, clogging of the well bottom can be assumed and measures should be taken to remedy the problem.

5.5. Limitations of the experiment for further considerations

In our experiment, the design of RRWH-SW was intentionally kept simple, in order to investigate the problems that might occur during the operation of these systems and observe its environmental effects.

On the other hand, using first flush removal devices, sedimentation tanks and filters can significantly improve water quality, especially considering the amount of sediment and organic matter load to the well (Dillon and Barry, 2005; Barry et al., 2013; Wang et al., 2015; Dillon et al., 2016; Soni et al., 2020). The use of these complementary settings is highly recommended during the implementation of real systems. Furthermore, using water flow meters would help determining the exact amount of water reaching the infiltration well from the gutters and thus it would enable a more accurate estimation of the efficiency of water harvesting.

The selected monitoring frequency of 30 min was adequate to investigate the efficiency of the system. However, in order to better understand the infiltration process during a specific infiltration event, especially in drier periods (i.e., summer), more frequent measurements are needed. Seasonal monitoring of water chemical parameters gave information on water quality changes both in SW and in P1–P2. In the case of SW, the water quality improvement was observable from the beginning of the experiment; however, these changes were not yet visible in P1–P2. Additionally, the sampling of well bottom sediments for total element concentration would give insight on the sedimentation and adsorption of elements during the infiltration process. For these reasons, the experiment is still ongoing in order to observe the long-term water chemical changes induced by rooftop rainwater infiltration (in the monitoring wells too), to rule out any possible contamination effect and thus gain acceptance and trust of the residents regarding RRWH-SW systems.

Water balance calculations (Yousif, 2022) suggest that the town-scale potential of RRWH-SH is quite remarkable. In order to determine the effect of town-scale rainwater harvesting more accurately, detailed geological and hydrogeological investigation of the area and 3D numerical modeling studies are needed. To delineate possible sites for new infiltration structures, suitability mapping could be a useful tool, either on regional or local scale (Adham et al., 2016).

5.6. Relevance and applicability of this research on settlement scale

The obtained results from the experiment can serve as a basis for numerical modeling studies for settlement-scale implementation of RRWH-SW in Kerekegyháza and in the Danube-Tisza Interfluve. This solution can provide a cheap, local scale and environmentally promising water replenishment method for the region suffering from serious water shortage. The experimental results, however, are not only applicable for the study region but can be used in other regions of the Earth as well.

There is a growing need worldwide for the development of municipal rainwater and stormwater management, from all sides (residents, municipalities, public utilities, water directorates, etc.) to avoid flash floods and related geoengineering problems, and to preserve water for droughts (Melville-Shreeve, 2017; Palla et al., 2017; Hussain et al., 2019; Tamagnone et al., 2020; Qin, 2020; Boroomandnia et al., 2021). Rainwater management is necessary from a protection point of view, but it is equally important to retain water and thus to adapt to climate change, to counteract overexploitation and to contribute to reaching sustainability (e.g., Pandey et al., 2003; Qi et al., 2019; Amos et al., 2020).

Socioeconomic acceptance and residents' participation is crucial for household level implementation of these systems and proper guidelines, policies and governmental subsidies can increase the willingness to harvest rainwater (Barthwal et al., 2014; Pawar et al., 2014). The results of this study and similar pilot projects (such as Pavelic et al., 2020 for basaltic terrains or Dillon and Barry, 2005 for borewells) can help to better understand the ongoing processes during rainfall events and promote the implementation of such projects worldwide.

6. Conclusions and recommendations

The 20-month-long monitoring of a field-scale rooftop rainwater harvesting and shallow well infiltration experiment can provide conclusions for the pilot area and can give basis for extending the application of this method to settlement level and using it in other parts of the Danube-Tisza Interfluve area. In addition, general conclusions can be drawn for similar projects in porous unconfined aquifers worldwide.

Direct conclusions for the pilot area:

1. *Quantitative changes and water level trends:* The total of 60.616 m^3 infiltrated precipitation, as a sporadic phenomenon, had little effect on the general decreasing water level trend observed in the monitoring wells of the pilot. The rate of water level decline on average was 0.28 m in the area (between Sept. 2020 and 2021). In the experimental wells (SW, P1, P2), 0.25 m, 0.35 m and 0.32 m water level decrease was observed in one year, respectively. On the other hand, by increasing the involved rooftop areas and continuing the infiltration could increase the quantitative potential of the method on settlement level: based on Yousif (2022), the water level increase can even reach 0.27 m y^{-1}, if the total collectable water amount of the roofs is infiltrated.
2. *Indication and troubleshooting of clogging:* The hydraulic conductivity of the aquifer originally was $1.8 \bullet 10^{-6}$ m s^{-1}. The well bottom sediments (after 15.5 months of infiltration) showed K values of $8.7 \bullet 10^{-7}$ m s^{-1} in the upper part and $3.9 \bullet 10^{-7}$ m s^{-1} in the lower part. By removing the accumulated sediment layer, the hydraulic conductivity of the well bottom increased to that of the aquifer.
3. *Diluting effect of infiltration on groundwater quality:* In the pilot, a significant improvement in water quality was achieved. Mg^{2+}, Na^{+}, Cl^{-}, SO_4^{2-}, NO_3^{-} concentrations and TDS decreased by 87%, 82%, 81%, 88%, 35% and 81%, respectively after the infiltration started and a further 96%, 93%, 68%, 58%, 96% and 35% decrease occurred in these parameters by the end of the monitoring period. This improvement could be maintained throughout the experiment and no significant increase occurred in these parameters.
4. *Trace element enrichment in rooftop-collected water:* Concentration of Zn, Sr, Cu, Mn, Ba and Al were 48.2, 6.9, 6, 4.3, 3.6 and 1.9 times higher, respectively, in roof runoff than in the precipitation. Despite the observed enrichment, none of them poses a considerable risk to groundwater quality. The only exception is Zn, whose concentrations in SW (114–249 μg L^{-1}) varied around the limit value for shallow groundwater (200 μg L^{-1}). However, its concentration decreased during the monitoring period and samples from the control wells also indicated lower values, which is a positive sign for long-term applications.

Conclusions for similar projects anywhere:

1. *Methodology*: The introduced experimental setup, namely a shallow (dug) well with two monitoring wells and involvement of further shallow reference wells with the monitored parameters can be adequate for groundwater quantity and quality evaluation regarding rooftop rainwater harvesting. The long-term (more than 1 year) monitoring could provide insight on not only seasonal but multi-annual processes and the effects of extreme precipitations which is significant to adapt to long-term water shortage.
2. *Parameters to follow quantitative effects*: The recharge rate in the infiltration well, water column changes, temporal changes of water level due to infiltration are applicable parameters for quantitative evaluation of the impacts in any study area.
3. *Efficiency of infiltration*: Continuous water level measurement in the infiltration well is an easy, relatively inexpensive and suitable way to monitor the efficiency of the process. Monitoring water column increase and comparing it with the calculated potential water column increase, as well as infiltration curve analysis after precipitation events can provide immediate information on the operation of the system.
4. *Maintenance:* The water column increase in the infiltration well can be the sign of clogging, thus it can be used for indicating the need for maintenance. If the ratio of ΔH and ΔH_{pot} is higher than 50–60% for 3 consecutive precipitation events, clogging of the well bottom can be assumed and measures can be taken to remedy the problem.
5. *Environmental tracers to monitor the pathways of infiltration*: Environmental isotopes are useful tracers to monitor the infiltration process, due the isotopic differences in precipitation and groundwater. Temperature is another useful parameter, as the temperature of precipitation is changing seasonally while that of groundwater is more stable. During the experiment, further natural tracers were found (Zn, U, B, Sr and H_2SiO_3) to monitor the pathway of infiltrated rainwater plume in groundwater as they have significantly different concentrations in SW and in P1–P2.

Based on the results and conclusions of the field experiment, the following recommendations are given for the implementation and monitoring of RRWH-SW systems on household level:

1. The materials and conditions of the rooftop and the gutters should be checked and if needed it should be renovated before implementation of RRWH-SW.
2. At least a filter mesh should be used in order to filter out larger contaminants (e.g., leaves). Further instruments, such as first flush devices and sedimentation tanks can be used to decrease the amount of sediment load to the well.
3. The well and the well bottom should be cleaned before infiltration. If possible, initial pumping of the well is advisable to freshen the water and to avoid initial contamination.
4. Proper maintenance of the gutters, filters and the well bottom, especially during and after autumn, is essential for maintaining the

efficiency of the system. Water column increase can help delineate clogging and indicate the need for maintenance.
5. Sampling of direct precipitation and roof-collected rainwater is necessary to delineate enrichment and possible contaminants. Further monitoring of critical parameters is advisable in the infiltration well.
6. If the continuous monitoring is not possible, at least seasonal water level and water quality measurements for main and critical elements are suggested to follow the infiltration process.

Preparing guidelines on how to properly implement, maintain and monitor RRWH-SW systems, taking into consideration national and European Union law regarding groundwater, could improve the effectivity of water retention and turn RRWH-SW from an unfavorable practice to a water management measure capable of improving the quantity and quality of water reserves.

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Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix

Table A.1

Measurements of main cations and anions in the shallow well (SW), in the monitoring wells (P1, P2) and in the control wells (KE01, KE02). The limit of quantification for sulfate is 25 mg L^{-1}, lower values are indicated with italics.

Well ID	Date [dd/MM/yy]	Ca^{2+} [mg L^{-1}]	Mg^{2+} [mg L^{-1}]	Na^{+} [mg L^{-1}]	K^{+} [mg L^{-1}]	HCO_3^- [mg L^{-1}]	Cl^- [mg L^{-1}]	SO_4^{2-} [mg L^{-1}]	NO_3^- [mg L^{-1}]	Hardness (CaO) [mg L^{-1}]	Calculated TDS [mg L^{-1}]
SW	14/01/20	63	215	117	6	795	148	279	120	583	1743
	31/01/20	32	28	21	1.6	114	28.1	33	78	109	336
	04/02/20	38.2	18.5	17	1.6	88	29	30	74	96	296
	27/02/20	22.9	16.2	6	2	63	11.2	*10*	70	69	201
	03/06/20	34.4	4.6	2	7	114	9	*4*	5	59	180
	10/09/20	70.6	3.5	2	3	177	1.1	52	1	107	310
	21/09/20	80	6.2	3	3	291	3.4	*9*	1	126	397
	15/12/20	22.6	22.6	2	6	74	4.5	*6*	1	34	139
	11/02/21	8.2	2.5	0.7	0.7	27	2.2	*7*	1	17	49
	24/03/21	26.7	2.5	1	2	74	6.8	*12*	3	43	128
	15/06/21	32.8	11.2	5	2	115	11.3	*19*	8	72	204
	21/09/21	51.3	1.2	1.5	4	135	9	*14*	3	75	219
P1	10/09/20	95.4	83.4	32	3	593	29.2	*8*	70	325	914
	21/09/20	90.3	89.6	32	3	731	25.5	41	30	333	1042
	15/12/20	110.8	89.6	37	4	704	36.1	62	40	361	1084
	11/02/21	92.3	83.4	35	2	650	38.2	64	70	321	1035
	24/03/21	88.2	88.4	36	2	677	36.1	64	50	327	1042
	15/06/21	90.3	99.6	44	3	711	38.4	92	50	355	1128
	21/09/21	90.3	117	47	2	629	49.7	133	100	396	1168
P2	10/09/20	91.6	81.1	42	4	593	32.6	49	70	315	963
	21/09/20	94.4	87.2	43	2	744	31.6	48	30	333	1080
	15/12/20	98.5	94.6	48	3	738	39.5	58	30	355	1110
	11/02/21	98.5	87.2	45	2	704	40.4	71	50	338	1098
	24/03/21	94.4	87.2	46	2	731	42.9	72	70	333	1146
	15/06/21	100.5	99.6	49	3	758	42.9	78	50	370	1181
	21/09/21	102.6	112.1	53	0.5	677	51.9	113	100	401	1210
KE01	21/09/20	22.6	5	3	11	122	2.3	*7*	0	43	173
	15/12/20	36.9	5	6.5	17	156	12.4	*11*	0	63	245
	24/03/21	73.9	23.7	25	17	338	22.6	57	3	158	560
	15/06/21	47.2	16.2	18	13	210	13.5	48	1	103	367
	21/09/21	43.1	3.7	9	15	149	11.3	29	3	69	263
KE02	21/09/20	20.5	1.2	1	1	68	3.4	*0*	1	32	96
	15/12/20	16.4	2.5	1	1	54	4.5	*6*	0	29	85
	24/03/21	26.7	1.2	2	1	68	9	*5*	8	40	121
	15/06/21	20.5	2.5	1	2	68	3.4	*4*	1	34	102
	21/09/21	18.5	2.5	2	2	54	9	*5*	3	32	96

Table A.2
Detailed laboratory measurements of water samples taken from the shallow well (SW), the monitoring wells (P1, P2), two control wells (KE01, KE02) precipitation collected directly in a bucket (PR) and rooftop rainwater collected from the tube system (RT). Limit values for groundwater are indicated based on Decree (2009), higher values are indicated with bold formatting. (Abbreviations: TSS – total suspended solids, EC – specific electrical conductivity, COD – chemical oxygen demand, DOC – dissolved organic carbon, TOC – total organic carbon, TDS – total dissolved solids, n.d. – below limit of detection, <x – below limit of quantification)

Date [dd/MM/yy]	PR	RT	SW				P1				P2				KE01	KE02	Limit value (Decree, 2009)
	14/04/21	14/04/21	11/02/21	24/03/21	15/06/21	21/09/21	11/02/21	24/03/21	15/06/21	21/09/21	11/02/21	24/03/21	15/06/21	21/09/21	21/09/21	21/09/21	
pH at 25 °C	5.92	6.73	6.91	7.43	7.53	7.29	7.42	7.48	7.45	7.32	7.4	7.44	7.31	7.36	7.44	6.81	
Total hardness [CaO mg L^{-1}]	0.81	7.39	13.8	38.5	56.5	73.9	336	322	335	392	346	339	349	349	71.7	34.9	
TSS [mg L^{-1}]	3.6	3.6	<2	74	37.6	87	925	1326	3879	3412	946	1196	3852	3382	2	4.4	
EC at 25 °C [μS cm^{-1}]	21	42	61	153	258	244	1219	1210	1283	1470	1319	1340	1388	1474	304	131	
COD [mg L^{-1}]	3.6	12.6	3.74	5.87	2.28	6.91	3.83	5.95	6.22	4.08	3.74	3.75	6.22	3.66	9.48	8.82	
DOC [mg L^{-1}]	1.3	8.1	1.9	–	–	–	3.5	–	–	–	3	–	–	–	–	–	
TOC [mg L^{-1}]	1.7	8.1	2.7	–	–	–	13	–	–	–	11	–	–	–	–	–	
Na^+ [mg L^{-1}]	0.93	1.04	0.72	1.5	2.54	0.86	41.6	42.2	48.8	66.6	60.9	62.9	68.3	62.5	5.68	0.69	
K^+ [mg L^{-1}]	0.6	2.1	0.89	1.91	1.97	2.6	2.01	1.57	2.18	0.97	1.32	1.37	2.4	2.02	14.8	1.66	
Ca^{2+} [mg L^{-1}]	0.48	4.63	8.84	24.8	28.5	47.8	95.1	84.1	83.2	97	95.6	90.1	93.8	91	39.1	22.8	
Mg^{2+} [mg L^{-1}]	0.06	0.39	0.63	1.62	7.12	3.01	87	87.7	93.5	110	90.8	91.1	93.4	95	7.25	1.28	
Fe^{2+} [mg L^{-1}]	<0.005	0.018	0.005	0.016	0.006	0.162	<0.005	<0.005	0.024	0.007	0.104	<0.005	0.006	0.007	0.042	0.005	
NH_4^+ [mg L^{-1}]	**0.54**	0.34	<0.1	<0.1	<0.1	<0.1	0.2	0.14	**1.52**	<0.1	<0.1	<0.1	**1.61**	0.13	<0.1	0.28	0.5
Mn^{2+} [mg L^{-1}]	0.0023	0.0088	0.0026	0.0179	0.0319	0.2336	1.1032	0.995	1.09	0.4377	0.1479	0.137	0.3457	0.4199	0.0396	0.008	
Total cations [mg L^{-1}]	2.61	8.53	11.1	29.9	40.2	54.7	227	217	230	275	249	246	260	251	66.9	26.7	
Cl^- [mg L^{-1}]	0.1	0.33	0.52	1.6	8.7	0.33	30.7	29.6	32.3	48.6	34.9	37.1	38.8	39.6	3.88	0.49	
NO_3^- [mg L^{-1}]	1.19	2.44	2.53	7.44	26.9	2.83	**82.7**	**81.3**	**86.8**	**165**	**86.8**	**95.9**	**87.1**	**103**	6.22	8.2	50
NO_2^- [mg L^{-1}]	<0.1	0.18	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	0.29	0.37	
HCO_3^- [mg L^{-1}]	6.1	15.25	12.2	60.4	59.8	150	702	628	659	622	714	665	726	622	144	54.3	
CO_3^{2-} [mg L^{-1}]	<3	<3	<3	<3	<3	<3	<3	<3	<3	<3	<3	<3	<3	<3	<3.0	<3.0	
PO_4^{3-} [mg L^{-1}]	<0.15	0.24	<0.15	0.19	<0.15	0.24	**0.58**	**0.52**	0.35	0.34	<0.15	0.37	0.32	0.16	**0.56**	**0.63**	0.5
SO_4^{2-} [mg L^{-1}]	1.1	4.05	8.38	10.8	16.5	6.77	53.5	55.7	67	133	61.1	61.1	54.6	97.7	25.2	9.21	250
F^- [mg L^{-1}]	<0.1	0.19	0.11	0.35	<0.1	0.11	0.36	0.36	0.44	0.46	0.33	0.1	0.38	0.38	0.15	0.11	1.5
Total anions [mg L^{-1}]	8.61	22.5	23.6	80.4	112	160	869	795	845	969	897	859	907	863	180	73.2	
H_2SiO_3 [mg L^{-1}]	<0.3	0.44	0.7	1.94	3.46	6.53	20.7	18.4	17.7	17.1	17.8	16.4	16.4	17.5	5.72	7.46	
TDS [mg L^{-1}]	11.2	31.5	35.4	112	156	221	1117	1031	1093	1261	1163	1121	1183	1131	253	107	
Li [μg L^{-1}]	n.d.	<2	3.22	5.81	2.41	4.67	9.73	9.23	5.64	10.6	12.3	9.81	5.74	11.1	10	<2	–
B [μg L^{-1}]	18.5	9.43	<5	16.2	n.d.	12.5	217	207	183	178	229	215	191	149	66.2	5.5	500
Al [μg L^{-1}]	6.85	12.8	13.5	10.9	14.7	252	8.71	4.25	19.7	17.9	256	4.07	7.32	13.7	39.7	24	–
V [μg L^{-1}]	<0.2	2.02	1.85	1.74	1.13	2.01	2.57	2.32	2.43	2.42	7.57	6.35	7.3	3.77	1.3	2.08	–
Cr [μg L^{-1}]	<0.5	<0.5	n.d.	n.d.	<0.5	<0.5	1.1	<0.5	<0.5	<0.5	1.76	<0.5	<0.5	n.d.	<0.5	1.19	50
Mn [μg L^{-1}]	2.27	9.85	3.08	19.9	34.9	197	1180	1070	1190	440	178	150	387	391	33.7	7.3	–
Co [μg L^{-1}]	n.d.	<0.2	<0.2	<0.2	<0.2	0.46	1.57	1.4	1.86	0.91	0.46	0.35	0.86	0.5	0.22	<0.2	20
Ni [μg L^{-1}]	n.d.	<1	<1	<1	<1	1.74	3.5	2.88	3.62	2.63	2.52	1.46	2.4	1.63	1.24	<1	20
Cu [μg L^{-1}]	0.63	3.79	2.13	3.48	2.86	6.18	9.98	5.53	2.28	4.56	4.14	4.15	2.08	5.18	3.97	11.3	200
Zn [μg L^{-1}]	6.01	**291**	**249**	**204**	114	185	21.1	48.3	24.9	18.7	9.76	16.1	17.4	6.17	69.4	38	200
As [μg L^{-1}]	n.d.	<1	<1	1.06	<1	1.39	2.71	1.79	1.9	1.37	1.16	<1	2.26	1.3	1.51	1.51	10
Se [μg L^{-1}]	<1	<1	n.d.	n.d	<1	n.d.	5.43	6.16	5.22	6.56	1.53	1.56	1.42	2.41	n.d.	n.d.	10
Rb [μg L^{-1}]	<0.2	1.41	0.54	1.67	1.43	2.37	0.56	0.5	0.81	<0.2	0.61	0.25	0.82	0.27	4.08	1.55	–
Sr [μg L^{-1}]	1.64	11.3	19.2	56.8	102	81.2	502	507	530	526	545	580	597	510	122	26.7	–
Mo [μg L^{-1}]	<0.2	<0.2	<0.2	0.28	<0.2	<0.2	2.34	2.23	2	2.31	1.33	1.46	1.46	1.65	0.23	0.56	20
Ag [μg L^{-1}]	n.d.	n.d.	<0.1	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	0.19	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	10
Cd [μg L^{-1}]	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	n.d.	<0.1	<0.1	<0.1	n.d.	<0.1	n.d.	n.d.	5
Sb [μg L^{-1}]	n.d.	<0.5	0.12	<0.5	0.15	0.24	0.3	<0.5	0.43	0.25	1.06	0.7	0.9	0.68	0.39	0.39	–
Ba [μg L^{-1}]	0.88	3.2	4.75	20.9	33.5	41.8	76.4	63.3	75.2	71.8	109	109	116	85.8	27.4	8.75	700

(*continued on next page*)

Table A.2 (*continued*)

Date [dd/MM/yy]	PR	RT	SW				P1				P2				KE01	KE02	Limit value (Decree, 2009)
	14/04/21	14/04/21	11/02/21	24/03/21	15/06/21	21/09/21	11/02/21	24/03/21	15/06/21	21/09/21	11/02/21	24/03/21	15/06/21	21/09/21	21/09/21	21/09/21	
Hg [μg L^{-1}]	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	1
Pb [μg L^{-1}]	<0.2	0.44	<0.2	<0.2	<0.2	0.45	<0.2	<0.2	0.25	<0.2	0.34	<0.2	0.22	<0.2	0.36	0.43	10
U [μg L^{-1}]	n.d.	n.d.	0.45	0.85	1.14	0.59	14	14.3	12.7	16.9	25.3	25	21.1	17.9	0.4	<0.2	–

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