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Enrichment of drinking water with Ca and Mg by a fluidized bed recarbonization reactor: a case study of Devičie, Slovak Republic

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ABSTRACT

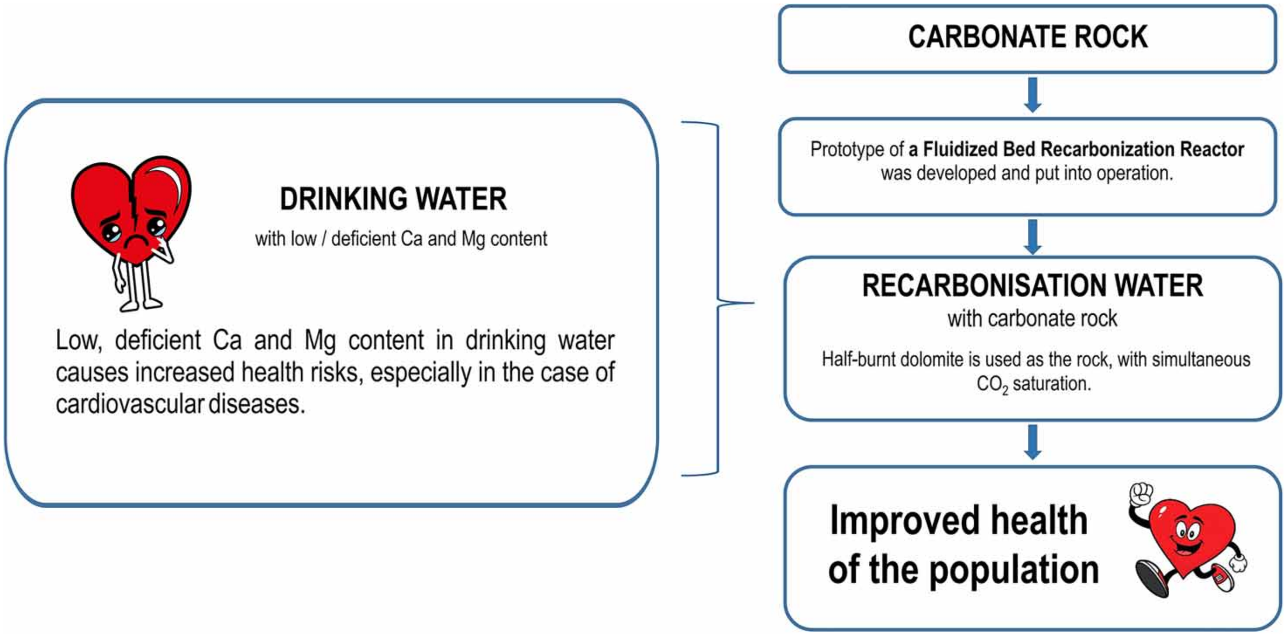
A low content of Ca and Mg in drinking water causes increased health risks. To increase the Ca and Mg contents in the drinking water supplied to the inhabitants of the village of Devičie, a prototype of a fluidized bed recarbonization reactor (RRF) was proposed and tested. A half-burnt dolomite (HBD) was used for the recarbonization. In the RRF, the HBD is kept in buoyancy with the help of water circulation. The capacity of the circulation pump is up to 5 $m^3 h^{-1}$ and the volume of discharged concentrate, which is added directly to the water source, is up to 0.2 $m^3 h^{-1}$. The volume of water circulated between the reactor and the circulating tank is many times higher than the volume of discharged water. In 24 h, the Ca and Mg contents stabilized at an equilibrium value of 80 mg L^{-1} for Ca and 120 mg L^{-1} for Mg, which corresponded to the equilibrium of formation and removal of ions from the system. The concentrate was diluted with the water in the reservoir at a ratio of 1:10, and it achieved the desired increase in Mg and Ca contents by more than 10 and 6 mg L^{-1}, respectively.

Key words: Ca, cardiovascular diseases, drinking water, Mg, recarbonization, recarbonization reactor

HIGHLIGHTS

- Increasing the Ca and Mg contents in soft drinking water is beneficial for human health.
- The prototype of the developed recarbonization reactor produces a concentrate with high Ca and Mg contents.
- The efficiency of the reactor is conditioned mainly by repeated washing of the carbonate rock.

GRAPHICAL ABSTRACT



1. INTRODUCTION

The drinking water supplied to the public must meet the limit values of the drinking water standards of the country concerned. In all cases, this sets the maximum permissible concentrations of various contaminants, whether of a biological or chemical nature. While all these substances, which are dangerous and harmful to human health, are very strictly regulated in the drinking water standards, very few standards assess the levels of biogenic (essential) elements, such as Ca, Mg, Na, K, F, I and so on. However, research conducted over the last 50–70 years has confirmed that Ca and Mg contents in drinking water have significant impacts on human health (Catling *et al.* 2005, 2008; Rubenowitz-Lundin & Hiscock 2005; Rapant *et al.* 2017; Rosborg & Kožíšek 2020).

When their contents are low, the incidence/mortality of cardiovascular diseases increases very significantly but also of oncological diseases, diabetes mellitus, digestive and respiratory diseases and other diagnoses. In addition, the life expectancy of people supplied with drinking water of low Ca (less than 30 mg L^{-1}) and Mg contents (less than 10 mg L^{-1}) tends to be 5 years lower than that of people supplied with drinking water of high Ca (>50 mg L^{-1}) and Mg contents (>25 mg L^{-1}) (Rapant *et al.* 2017, 2020, 2021). This unfavorable condition could be modified by increasing the Ca and Mg contents in drinking water, by the process known as recarbonization (RC). Different processes and different carbonate rocks are used for RC of drinking water, mostly under CO_2 saturation. In particular, flow-through systems are used where carbonate rock is added to a flow-through device based on various filters, through which the entire treated water flows (Haney & Hamann 1969; Al-Rqobah & Al-Munayyis 1989; Olejko 1999; Withers 2005; Luptáková & Derco 2015).

To enrich the drinking water, supplied to the inhabitants in the village of Devičie, with Ca and Mg, a prototype of a fluidized bed recarbonization reactor (RRF) was developed. In the RRF, a layer of solid particles of carbonate rock – half-burnt dolomite (HBD) – is kept in lift and in motion by the bottom-up flow of water, which intensifies the dissolution process of the carbonate rock. The aim of the present paper is to verify the functionality and efficiency of the manufactured RRF prototype for the water source of Devičie, which is used to supply drinking water to the local population, i.e. whether there is a sufficient enrichment of drinking water with Ca and Mg, respectively, and the hardness of drinking water. Enrichment of drinking water with Ca and Mg may reduce the potential health risk of local residents due to low Ca and Mg contents in drinking water. We expect a progressive improvement of people's health, which will be monitored by measuring arterial stiffness (Rapant *et al.* 2019a, 2019b).

2. AREA DESCRIPTION

The water source of Devičie (Figure 1) is used to supply the population of the village, approximately 300 inhabitants. Water is pumped into the reservoir from the hydrogeological borehole (37 m) according to the current water consumption. The average annual water consumption is approximately 11,000 m^3 and the daily consumption is approximately 30 m^3. The borehole is located in Neogene volcanics (andesites and their pyroclastics). Given the geological setting, the groundwater in Devičie is characterized by low total mineralization (approximately 320 mg L^{-1}), low water hardness and low Ca and Mg contents (Table 1). The health status of the population of the village Devičie is significantly worse compared to the average of the Slovak Republic (Rapant *et al.* 2019a, 2019b).

However, the Slovak drinking water standard regulates Ca and Mg contents and water hardness (expressed as Ca + Mg in mmol L^{-1}) only as recommended values. Table 1 shows that the values of the above-mentioned parameters are at the lower end of the regulated recommended values. Based on the risk analysis, an increased health risk from Ca and Mg deficiency and low hardness of water for the local population has been confirmed (Rapant *et al.* 2019a, 2019b, 2020, 2021).

The mean hazard quotient level for deficient elements (HQ_d) was found to be 1.6 for Ca, 1.56 for Mg and 1.58 for water hardness. To eliminate the above risk, Ca content should be increased at least by 5 mg L^{-1}, Mg content by 6 mg L^{-1} and water hardness by 0.35 mmol L^{-1}. We have set ourselves the objective to increase Ca content by 6 mg L^{-1}, Mg content by 10 mg L^{-1} and water hardness by 0.5 mmol L^{-1}.

Figure 1 | Map showing the location of Devičie.

Table 1 | Ca, Mg and water hardness contents of drinking water in Devičie compared to the standard values of the Slovak drinking water standard and proposed concentrations of Ca, Mg and water hardness after recarbonization

	Ca	Mg	Water hardness
	[mg L^{-1}]		[mmol L^{-1}]
Devičie	30.0	10.2	1.15
Slovak drinking water standard[a]	>30	10–30	1.1–5.0
Proposed values after RC[b]	35–40	16–21	1.5–1.6

Note: [a]Decree of the Ministry of Health No. 247/2017 Coll.
[b]The proposed values are based on a risk analysis.

3. MATERIALS

3.1. Carbonate rock leaching

To enrich drinking water as a rock material, we use the HBD, which is characterized by the highest solubility of carbonate rocks (Tuček *et al.* 2017). The HBD is produced from dolomite during annealing at 600–800 °C. Upon annealing of dolomite [CaMg $(CO_3)_2$], magnesium changes to form oxide and calcium remains as carbonate. Magnesium oxide is much more soluble than Mg carbonate, and therefore, Mg ions are preferentially released when the HBD is dissolved in the liquid phase. We used the HBD by Magno-Dol (Germany) with the size fraction of 2.0–4.5 mm. Magno-Dol is EU approved for drinking water treatment. It contains approximately 98.2% of Ca and Mg oxides and carbonates, approximately 0.9% of Si, Al and Fe oxides and 0.8% of water. Other elements are less than 0.1%. Toxic heavy metals are present only in trace amounts. Food CO_2 (Messer Tatragas) as an oxidizing agent for dissolving the HBD, which contains more than 99% CO_2 and is approved by the EU for food purposes, was used.

4. METHODS

4.1. Design of a RRF

To enrich drinking water with Ca and Mg and for the specific conditions of the water source, Devičie designed a RRF (Figure 2).

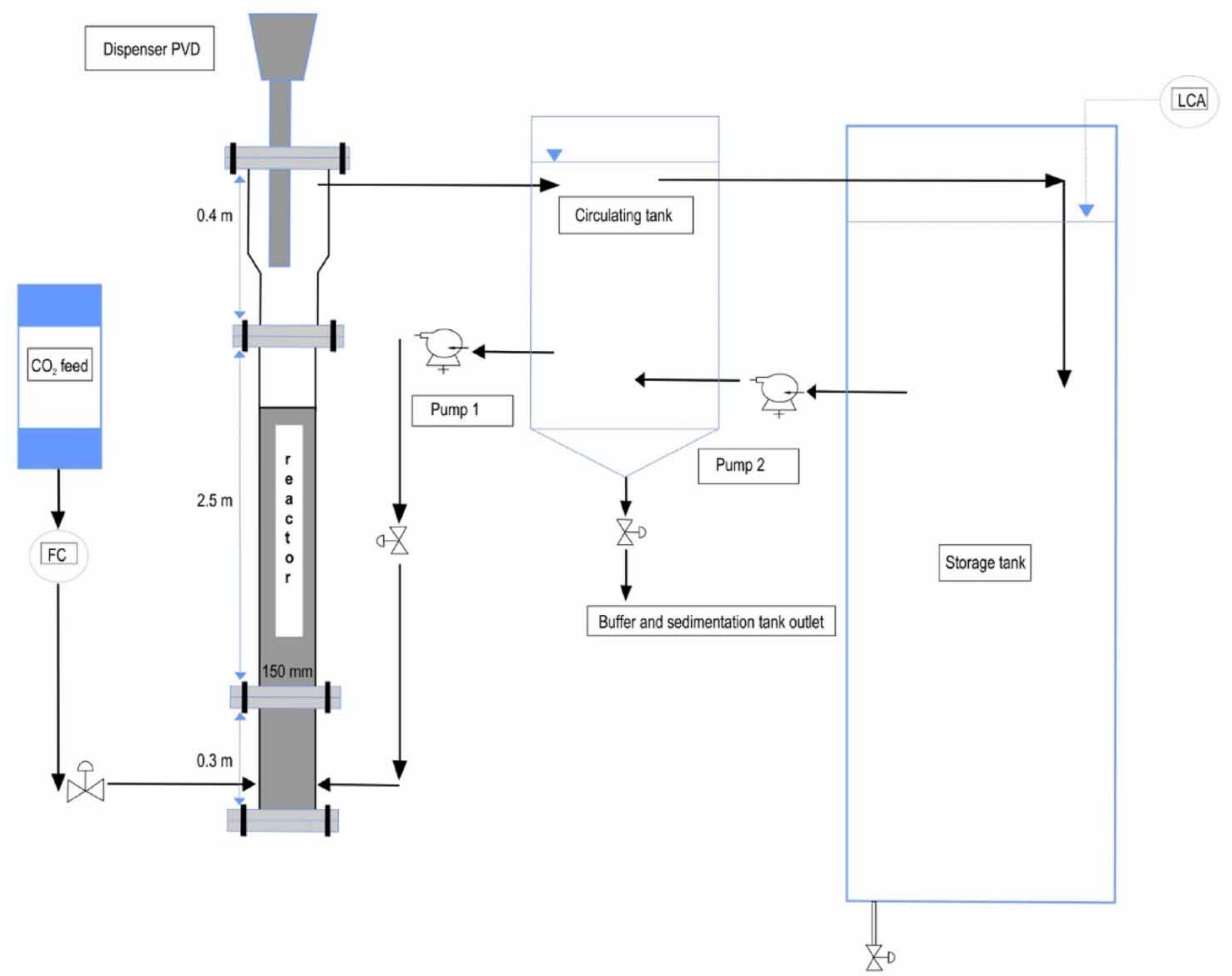


Figure 2 | Schematic diagram of the drinking water recarbonization plant at the water source in the village of Devičie.

The whole device has three main parts: the reactor body, the circulation tank and the dosing tank, for more details see Jelemenský (2020).

The system is driven by two pumps. One pump serves as a circulation pump (circulation between the reactor and the circulation tank) and the other for the discharge of the produced concentrate to the drinking water reservoir. The carbon dioxide is supplied by pressurized bottles with serial connection. The RRF is approximately 3.3 m long and 150 mm in diameter. The top section is extended and has a diameter of 250 mm. An HBD doser is located at the top of the reactor. The reactor contains 20–40 kg of the HBD. The settling circulation tank has a capacity of approximately 0.5 m^3. The circulation pump has a capacity of up to 5 m^3 h^{-1}. The recharge pump has a capacity of up to 0.2 m^3 h^{-1}. The output of the circulation pump is therefore approximately 25 times higher than that of the make-up water pump, thus ensuring multiple washing of the HBD and thus a higher Ca and Mg content in the concentrate. The produced concentrate is fed by gravity directly into the water reservoir where it is mixed with the drinking water. Carbon dioxide is fed into the bottom of the reactor and its flow is measured by a rotameter. The whole plant is made of materials that resist corrosion. All materials used comply with the hygiene requirements for contact with drinking water.

The design and location of the reactor obey the existing conditions of the water reservoir, and the reactor has been positioned in such a way that no structural modifications are required. Photodocumentary is available in LIFE WATER and HEALTH (2018).

4.2. Semi-operational test

Prior to the commissioning of the RRF to the water source of Devičie, a semi-operational test was carried out at the reactor in laboratory conditions. The first series of tests was aimed to determine the functionality of the plant in order to identify possible limitations in terms of HBD quantity, water circulation rate, CO_2 consumption and water inflow to the system (Jelemenský *et al.* 2019).

In the first step, the system was filled with 10 kg of HBD and 0.45 m^3 of water. The circulating water flow rate was 3.6 m^3 h^{-1}, which corresponds to the fluidization threshold. The CO_2 inflow was 1 nL CO_2 min^{-1} and the pressure at the regulator was 1 barg. Conductivity, pH, water temperature and Ca and Mg contents were monitored. After 8 h of RRF operation, the conductivity increased from 550 to 1,400 μS cm^{-1}, the pH decreased from 7.75 to 6.22 and the water temperature increased from 17.0 to 22.8 °C. The Ca content increased from 57.1 to 67.2 mg L^{-1} and the Mg content increased from 20 to 160 mg L^{-1} (Figures 3 and 4). Then, additional 10 kg of HBD were added and the conductivity increased to 4,756 μS cm^{-1}, and Ca and Mg contents increased to 104.5 and 665.0 mg L^{-1} within the next 27 h, respectively (Supplementary Material, Figures S1 and S2). Then, water was added in the reactor with an influent flow of 316 L h^{-1}. The water circulation and CO_2 influent were not changed. The freshwater influx showed a significant dilution. After 6 h, the Ca and Mg contents stabilized at approximately 60 and 75 mg L^{-1}, respectively. Similarly, the conductivity value decreased after approximately 6 h to 600 μS cm^{-1} (Supplementary Material, Figures S3 and S4). After reducing the inflow of freshwater from 316 to 60 L h^{-1}, the conductivity of the water increased significantly to more than 3,100 μS cm^{-1}, and Mg and Ca contents increased significantly to more than 400 and 85.3 mg L^{-1}, respectively. The concentrate was opaque water with rock microparticles. Therefore, the water inflow was adjusted to 120 L h^{-1}. This flow in the real system meant mixing the concentrate and water to the consumer in a ratio of 1:10. The change in freshwater inflow from 60 to 120 L h^{-1} was manifested by a decrease of Mg concentration from 400 to 235 mg L^{-1}, Ca concentration from 85.3 to 71 mg L^{-1} and conductivity from 3,100 to 1,825 μS cm^{-1} (Figures 5 and 6). These values can be considered stable. The decrease of Mg concentration to 131 mg L^{-1} between 114 and 122 h was due to loss of CO_2 in the system. At 118 h, the CO_2 flow was restored. Furthermore, several tests were carried out under different conditions, varying the water flow rate, the amount of CO_2 supplied and the amount of the HBD (Jelemenský *et al.* 2019).

The results were similar. With decreasing water inflow, Ca and Mg production increased, and a similar dependence was observed with increasing CO_2 inflow although not very significantly. The optimal inflow and outflow of water in a real system is determined to be 120 L h^{-1}. This inflow and outflow in the real system meant mixing the concentrate into the water in the reservoir at a ratio of 1:10. Mixing the concentrate with the water in the reservoir results in an increase of Ca concentration by 6.4 mg L^{-1} and Mg concentration by 12 mg L^{-1}.Based on the average annual water consumption in the village of Devičie (11,000 m^3 $year^{-1}$) and the required increase of Mg content by 10 mg L^{-1}, the daily Mg requirement is 301 g. The obtained daily values of Mg produced under the different conditions used are given in Table 2.

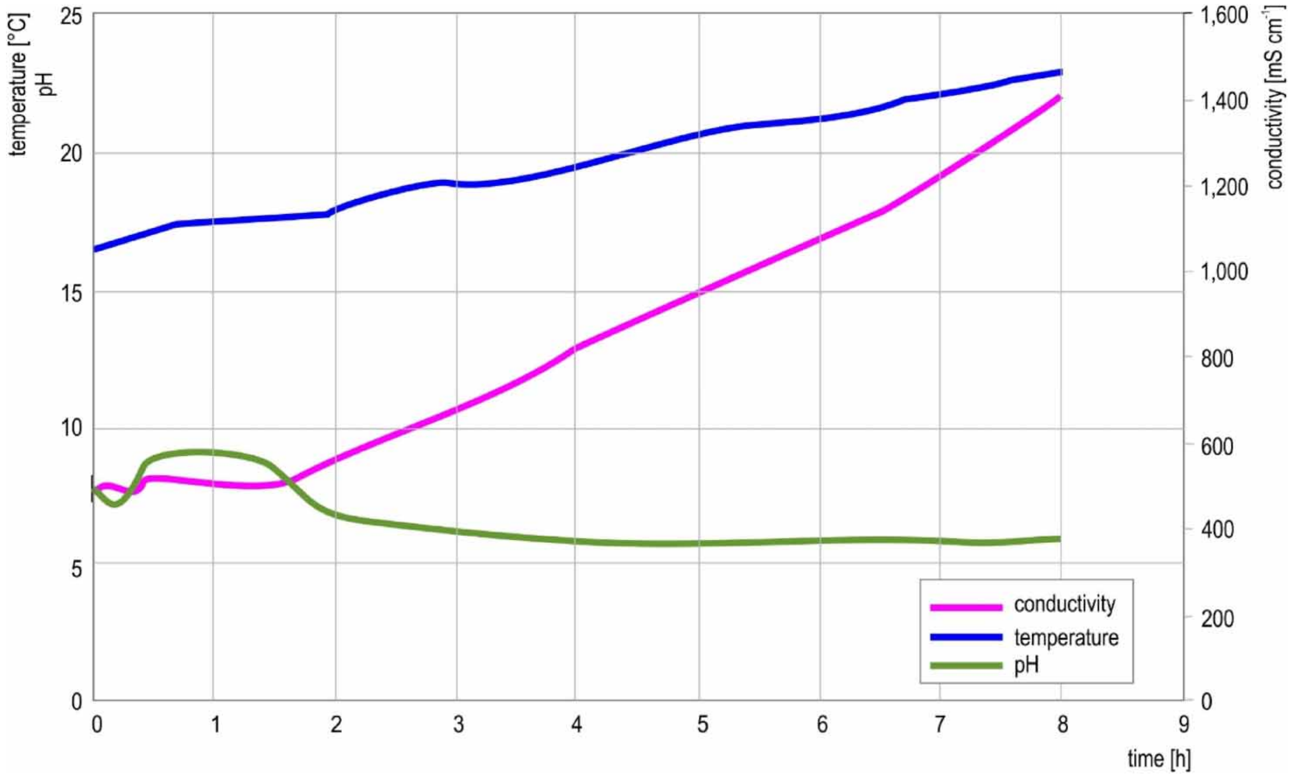


Figure 3 | Conductivity, pH and temperature at the beginning of saturation.

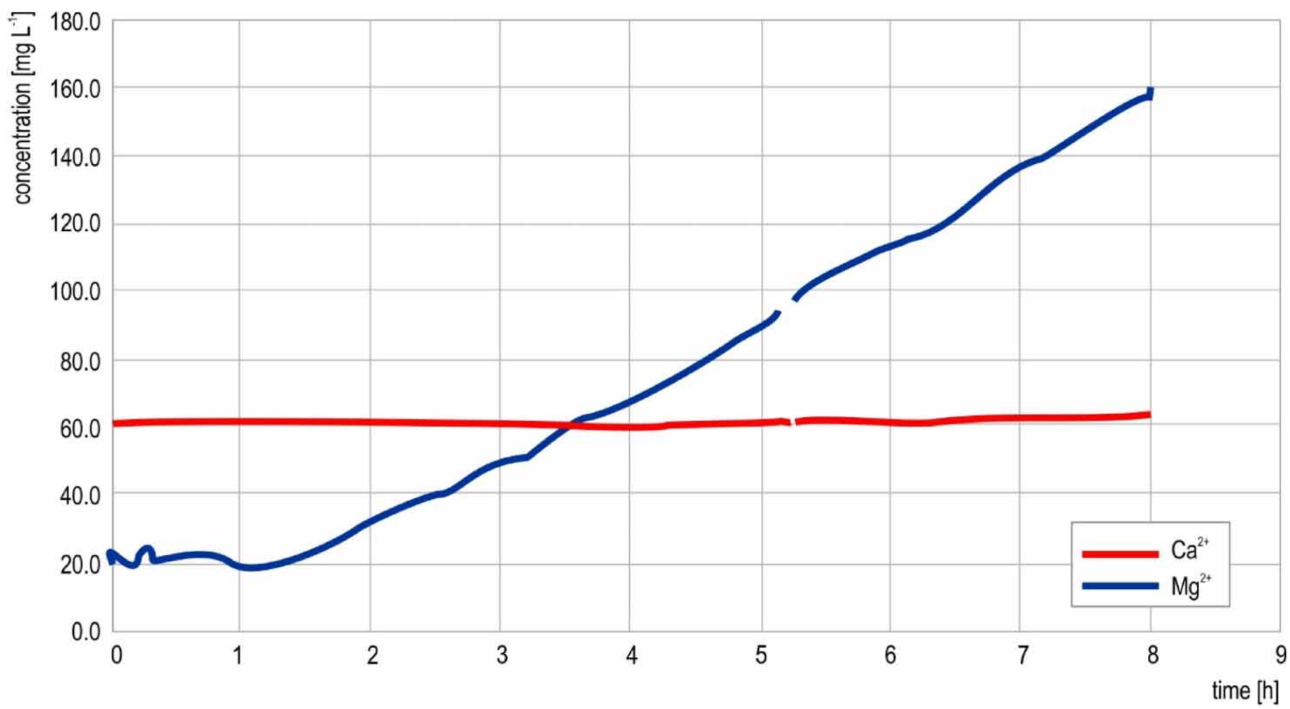


Figure 4 | Concentrations of Ca and Mg at the beginning of saturation.

It is clear from Table 2 that for all RC conditions used, the amount of Mg produced exceeded considerably the requirement of 301 g day^{-1}. A further increase in Mg production can still be assumed after dissolution of HBD microparticles in the water in the reservoir as the concentrate contained a residual amount of unconsumed CO_2 at 450 mg L^{-1}. However, a gradual

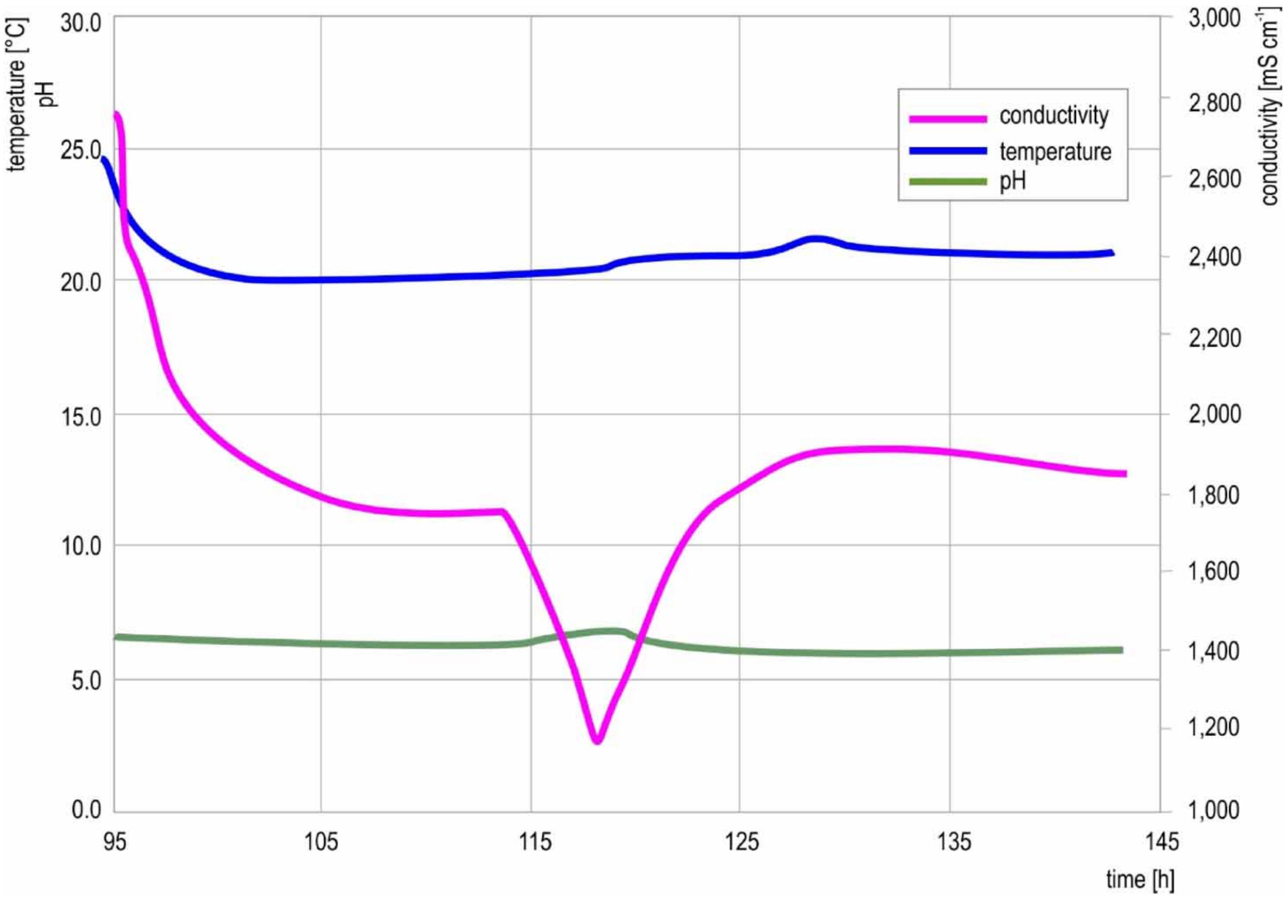


Figure 5 | Conductivity, pH and temperature after changing the water flow from 60 to 120 L h^{-1}.

decrease of Mg and increase of Ca content with increasing time should be expected. Once the MgO has dissolved, $CaCO_3$ starts to dissolve at an increased rate. Additionally, it should be taken into account that at lower temperatures in a real-water reservoir system (more than 10 °C), the solubility of CO_2 is higher and dissolution will be more intense. An assumed HBD and CO_2 consumption of approximately 500–600 kg and 600–700 kg, respectively, was determined as the maximum consumption of HBD and CO_2 for the increase of Mg content by at least 10 mg L^{-1} and Ca content by at least 6 mg L^{-1} for the average annual water consumption of 11,000 m^3. Consumption of the HBD was based on 65% utility of Mg and Ca. However, it can be assumed that the utility of Mg and Ca will be higher and HBD consumption will be lower. The proposal of operating conditions of the water RC process in the water source of Devičie, with an average daily water consumption of 30 m^3 day^{-1} and an increase of Mg content by at least 10 mg L^{-1} and Ca by at least 6 mg L^{-1} is the following:

- water circulation rate between the reactor and the storage tank: 3.8–4.1 m^3 h^{-1},
- water flow to the tank: 110–130 L h^{-1},
- achieved concentration of dissolved Mg: 100–130 mg L^{-1},
- CO_2 flow rate: 58.5 g h^{-1} (2 nL min^{-1}),
- HBD allowance per day: 6 kg.
- Mg increase: 12.56 mg L^{-1}
- Ca increase: 6.05 mg L^{-1}

5. RESULTS

The RC reactor was put into operation on 25 June 2021 under the following conditions. The circulating rate of water was 3.8 m^3 h^{-1} and the CO_2 inflow was 2 nL min^{-1}. Approximately 25 kg of HBD was poured into the reactor. The water

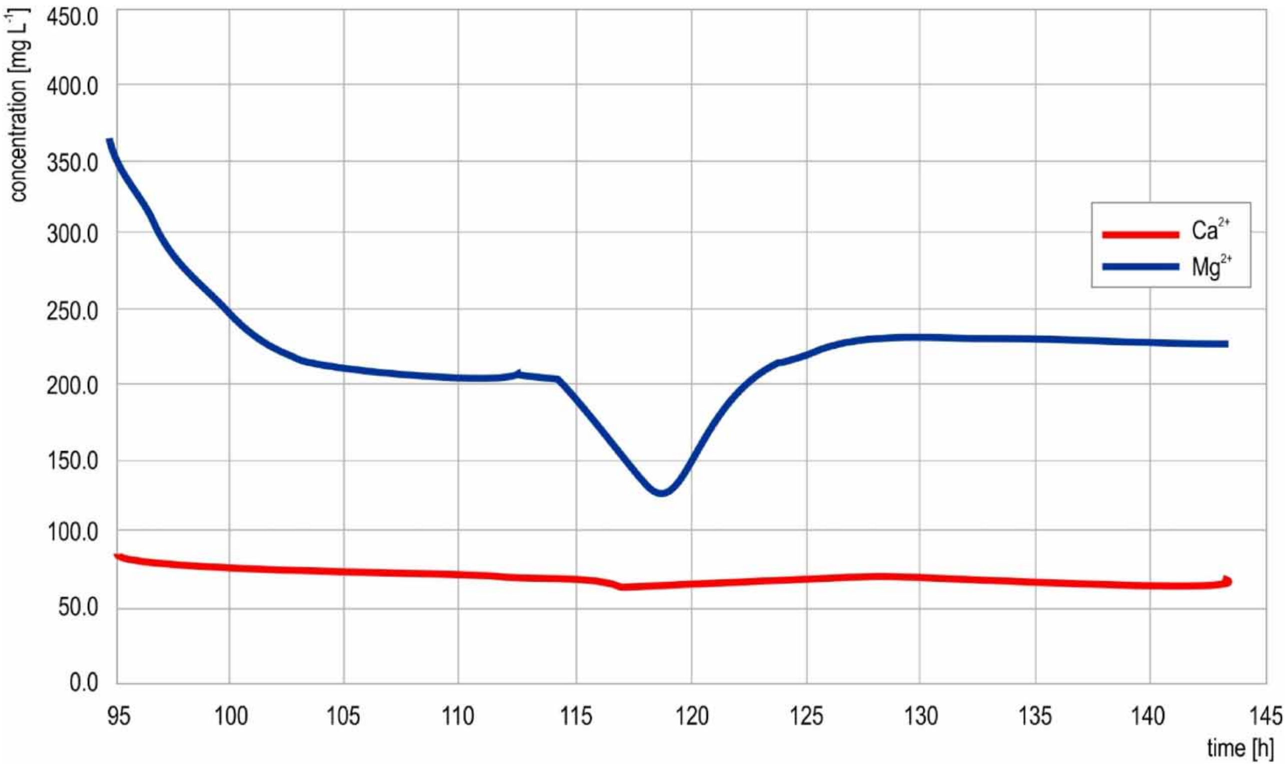


Figure 6 | Concentrations of Ca and Mg after water flow change from 60 to 120 L h^{-1}.

Table 2 | Steady state magnesium production at different process conditions

Water inflow (L h^{-1})	60	120	120	180	316
CO_2 flow (nL min^{-1})	1	1	2	2	1
Magnesium production (g day^{-1})	564.5	567.3	633.6	695.5	455.1

inflow to the reactor was 120 L h^{-1}. The results for the first 60 days of reactor operation are shown in Table 3 and for Mg and Ca concentration in Figure 7. Since the required content of Mg (120 mg L^{-1}) and Ca (80 mg L^{-1}) in the concentrate in the circulating tank was recorded in 48 h, CO_2 inflow was gradually decreased to 0.1 nL min^{-1} (30 g h^{-1}).

6. DISCUSSION

It is clear from the results of the semi-operation test and 60-day reactor operation in the village of Devičie (Table 3) that the required increase of Ca (at least by 6 mg L^{-1}) and Mg contents (at least by 10 mg L^{-1}) was achieved using RC of drinking water in Devičie. A conductivity of 1,245 μS cm^{-1} and Ca and Mg contents (80.4 and 126.2 mg L^{-1}, respectively) in the circulating tank were obtained in 48 h. The first increase of Ca and Mg contents in drinking water in the village of Devičie was recorded after 5 days. The conductivity value increased from 298 to 360 μS cm^{-1}. After about 10 days, when the conditions in the water pipeline stabilized, the required increase of Ca and Mg contents in the drinking water supplied to the population in the village of Devičie was achieved. This condition was maintained throughout the system for the first 2 months since the reactor has been put in operation. The reservoir in Devičí has a volume of about 90 m^3. After the depletion of approximately 10 m^3, water began to flow automatically from the borehole into the reservoir. Therefore, at the time of adding freshwater to the reservoir, the Ca and Mg contents decreased slightly. It was evident from the obtained results that the production of dissolved Mg, Ca ions and water hardness by the designated RRF prototype was fully assured.

Table 3 | Results of water recarbonization in the circulation tank and in the water in the village

	Circulation tank				Water in the village			
Day	Conductivity [µS cm^{-1}]	Mg [mg L^{-1}]	Ca	pH	Conductivity [µS cm^{-1}]	Mg [mg L^{-1}]	Ca	pH
0					298	10.2	30.1	
1	1,245	126.2	80.4	6.4	300	10.1	30.1	7.1
3	1,250	134.1	84.8	6.5	310	10.4	30.3	7.3
5	1,190	128.3	82.5	6.4	360	15.2	32.6	7.2
10	1,140	115.9	82.4	6.5	380	20.7	33.7	7.1
15	1,180	118.3	82.3	6.5	425	21.4	35.8	7.1
30	1,180	119.4	83.2	6.4	430	21.5	36.8	7.2
40	1,067	116.5	84.2	6.5	428	21.0	37.1	7.1
50	1,098	118.2	85.1	6.5	472	21.2	37.1	7.1
60	1,105	119.4	85.4	6.4	444	21.2	37.4	7.2

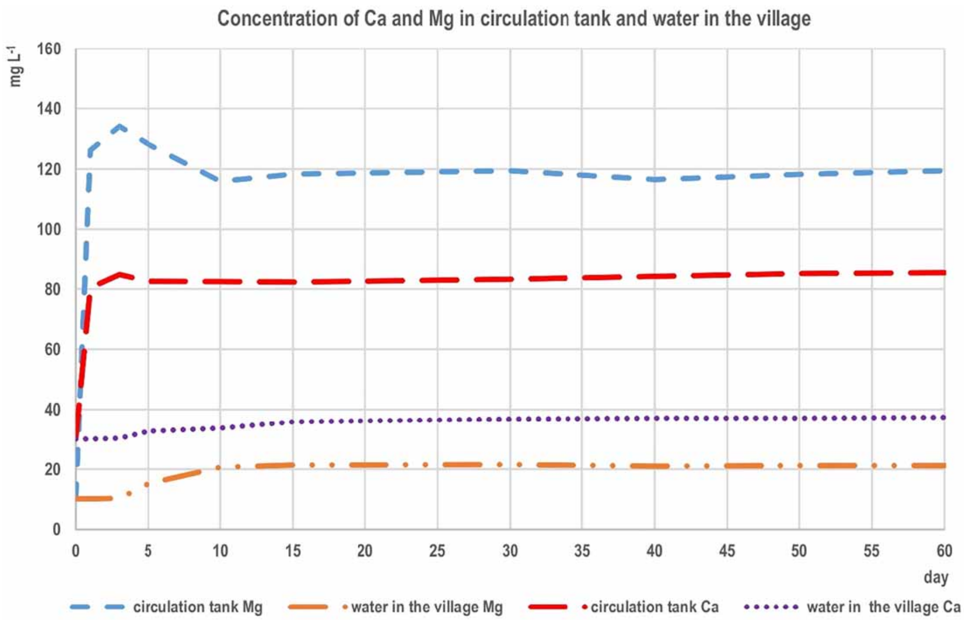


Figure 7 | Concentrations of Ca and Mg in circulation tank and water in the village.

Similar RRFs can be used for water sources with yields up to 5 L s^{-1}. However, it is necessary to increase the diameter of the reactor (up to 0.4 m), the volume of the circulating tank (5–10 m^3) and the flow rate of the circulating water (min to 15 m^3 h^{-1}). Such a larger RRF is currently being prepared within the LIFE – WATER and HEALTH project (LIFE WATER and HEALTH 2018) for the water sources Kokava nad Rimavicou with an average yield of 3.3 L s^{-1}. The obtained results demonstrated the robustness of the equipment over a wide range of conditions. The amount of Ca and Mg ions produced, and their ratio can be controlled by the flow rate of circulated water, by the amount and age of the HBD and the flow rate of CO_2.

It is very difficult to compare our results of the RC process with similar works in the world literature. On one hand, to our best knowledge, it is the first developed RRF. The reactor was developed and manufactured within the solution of the LIFE project – WATER and HEALTH (https://fns.uniba.sk/lifewaterhealth/). The flow-through system is currently used in several countries. However, water companies practically do not publish the results, they consider them as their know-how, or they are protected by patents. No data are available on the consumption of rocks, the quantities of CO_2 required, etc. In Slovakia, several semi-operational trials of the RC process have been carried out in water supply reservoirs for drinking water with low hardness and Ca and Mg contents (Barloková *et al.* 2017).

For example, the water hardness increased from 0.4 to 1.2 mmol L^{-1} in the water reservoir of Hriňová using the HBD and CO_2 saturation by flowing water through the filter, and the increase of water hardness from 0.35 to 1.52 mmol L^{-1} was obtained in the water reservoir of Turček in semi-operational trials. However, only 0.05 L s^{-1} of water was recarbonized.

RC of drinking water with low hardness or desalinated seawater is currently being implemented in several countries around the world (e.g. Scotland, Sweden, Spain and especially Arab countries). From the data available to us, desalinated seawater in Sweden is being mineralized in this way at Sandvik Oland and Gotland (Swedish Royal Court 2017; Bygga bo och miljö 2020). The amount of recarbonized water is 3,000 m^3 day^{-1} at Sandvik and 7,000 m^3 day^{-1} at Gotland. The target values for RC are 30 mg L^{-1} for Ca and 10 mg L^{-1} for Mg. The HBD is used as the rock at the Sandvik site and calcite at the Gotland site. In both cases, CO_2 saturation is used. Further data are not available.

7. CONCLUSIONS

Our RRF has demonstrated high reliability and robustness over a wide range of conditions. The amount of Ca and Mg ions produced can be effectively controlled mainly by the flow rate of circulated water, amount of the HBD and the flow rate of CO_2. The production of dissolved Ca and Mg is mainly ensured by the intensive circulation of water between the reactor and the circulation tank. Under the given conditions of the water source of Devičie (source capacity of 0.35 L s^{-1}), the increase of Mg content by 10 mg L^{-1}, Ca content by 6 mg L^{-1} and hardness by 0.5 mmol L^{-1} in drinking water can be safely achieved using the RRF. The RRF developed by us is relatively simple and inexpensive. It does not require any structural modifications to the existing water reservoir. An area of 2 m^2 is required for its placement. Such type of reactor can be used to recarbonize water with a yield of up to 5 L s^{-1}. The operation of the RRF is also inexpensive. For water treatment at the water source in the village of Devičie (11,000 m^3 $year^{-1}$), the HBD consumption is approximately 500–600 kg and the CO_2 consumption is approximately 600–700 kg. The reactor efficiency, i.e. the production of Ca and Mg ions over the next 5 years of operation, will be available on the website http://uniba.sk/lifewaterhealth. The expected positive effect of enriched drinking water on the cardiovascular system of the population of the village of Devičie will be monitored at half-yearly intervals through arterial stiffness.

AUTHOR CONTRIBUTIONS

S.R. and V.C. conceptualized the study; P.Č. and V.C. prepared and wrote the original draft; reviewed and edited the article; S.R. visualized the study, designed the reactor, prepared the methodology of semi-operational test. All authors have read and agreed the article to be published.

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INFORMED CONSENT STATEMENT

Informed consent was obtained from all subjects involved in the study.

DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

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