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Isotopic tracing and quantitative assessment of rainwater in a separate sewer system in Tiszavasvári, Hungary

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ABSTRACT

The hydrological processes that commonly occur within the sewer system i.e., rainwater percolation or groundwater infiltration, can be easily described applying water isotopes. This study investigates the potential of tritium and stable isotope composition (δ^2H and δ^{18}O) of water for tracing rainwater entering urban sewer and estimating the amount. The work consists of collecting and analyzing wastewater and rainwater samples over a six-day-long period including a 30-min-long rainfall event, and then applying the isotopic results to determine the mixing ratios in the sewer pipes and the sensitivity of the applied tracing methods. Results reveal that the isotopic composition of wastewater reacts to inflowing rainwater sensitively: detectable flow rate increase resulting from rainfall is 0.62 m^3h^{-1} for tritium, 2.49 m^3h^{-1} for δ^{18}O and 1.25 m^3h^{-1} for δ^2H. While applying variation of discharge, these values are 12.15 m^3h^{-1} and 18.91 m^3h^{-1} in daytime and nighttime, respectively. Due to the more positive stable isotope values of precipitation in the summer half-year, isotopic difference of rainwater and groundwater (being less enriched) is more notable, subsequently, traceable amount of parasitic rainwater in case of the examined area is lower than in the winter half-year. In case of groundwater recharged under cold climate conditions sampling should be carried out in summer in the night hours, after an at least three-day-long dry period. Though, all three isotopic tracers applied in this study proved to be suitable to detect small amount of parasitic precipitation, tritium came out as the most sensitive one.

1. Introduction

Urban sewer systems are one of the most important water infrastructures all over the world. Functional efficiency and structural quality of them are crucial to ensure safe transportation of domestic and communal wastewater to treatment plants without infiltration [1]. There are two basic constructions of sewer systems for transporting foul wastewater: combined and separated systems. In contrast to separate sewage disposal, in combined systems wastewater and precipitation water (rain, snowmelt) are drained in common mains in the same pipe system. This may drastically increase the costs associated with sewage treatment and lead to environmental problems due to flooding of urban areas in case of extreme rain events [2]. According to estimations reported in the literature [3,4], the proportion of dry-weather extraneous waters is about one third of the daily inflow, which demonstrates the magnitude of the problem. Therefore, rainwater inflow in a sewer network is a major concern in urban water management. Recent urban water planning guidelines suggest revised rainwater management strategies, which call for rainwater source control and rainwater re-infiltration [5]. [6] state that owing to an increase in surface runoff overflow due to climate change, the sewer system in the study region has become more loaded due to flooding. They found that even cracked manhole covers and gaps may allow significant amount of stormwater to leak into the system.

According to [7], the waters that should not be present in sewers may be sorted into four groups: 1) groundwater in combined sewers, 2) waste water in the separate storm sewers, 3) domestic waters leaking from supply pipes and entering the combined sewers, 4) rainwater in the separate sewers. This paper focuses on the fourth type of extraneous water aiming to trace rainwater illegally drained into the separate sewer network.

Although the construction is more difficult and expensive, operation

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of separate sewer systems is a cost effective solution to reduce overloading of sewers. Draining and transporting of precipitation (storm sewer) and wastewater (sewer) separately in two distinct pipe networks reduce the hydraulic loading and the possibility of overloading of the sewer systems, which decreases the probability of local flooding and deterioration of urban infrastructures [8]. Additionally, it leads to lower maintenance costs and energy consumption in treatment plants. A defect of this solution is the occurrence of improper connections when rainwater outlets to the separate sewer system [2,9] resulting in a number of undesirable consequences on the performance of the wastewater system including the treatment plant. Sewer systems are not designed to handle large amounts of storm water and the consequences of a heavy rainfall are similar to those occurring in the case of overloading of combined sewers. Furthermore, the dilution of wastewater with unwanted (parasitic, extraneous) rainwater has its negative effects on treatment plant capacity and efficiency [10]. Moreover, under favorable conditions, through cracks and loose joints in sewer pipes, a significant amount of groundwater may infiltrate into the system further increasing the volume of extraneous water [9,11,12]. Infiltration of groundwater and inflow of storm water may also escalate pipe ageing and increase the endangerment of adjacent infrastructure. The positive effects of infiltration and inflow (I/I) include higher total flow rate, improved flushing, lower concentrations and decrease of sewage temperature resulting in increased transport capacity and reduced sediment build-up, anaerobic processes, smell development and corrosion [8].

Assessment and detection of I/I in sewer systems are critical issues in the long-term urban water management and several methods based on various markers have been proposed to fulfill these requirements [2,12] including flow rate measurement, smoke testing, dye testing, DTS (distributed temperature sensing), tracer methods, etc. [12]. According to [13] an ideal marker should be unique, easily analyzable and have a conservative behavior, conditions that are difficult to be fully satisfied. Reviewing the suitability of several chemical species as markers (including major and minor ions, metals, CFCs, microbiological parameters, stable isotopes, colloids, etc.) for indicating recharge sources, [13] concluded that there are no ideal markers and emphasized the need for a multi-component approach rather than using individual markers. [4] claimed, that in sewer systems, it is typically difficult to find suitable inherent markers since sewage chemistry usually exhibits substantial background fluctuations that conceal the natural tracer signal.

In this study water isotopes ($\delta^{18}O$, $\delta^{2}H$ and ^{3}H) have been applied to assess rainwater inflow into the sewer system. Water stable isotopes have long been used as tracers in hydrological studies to qualify or quantify recharge and discharge conditions or physico-chemical processes impacted on the studied water bodies [14–18]. They are also relevant in assessing the relative proportion of two or more water sources [19]. These methods require only a distinct isotopic composition of wastewater and infiltrating groundwater and/or inflowing precipitation. [4] demonstrated that stable isotopes of water are promising tracers for monitoring both the origin and the amount of extraneous waters from two sources (drinking water and groundwater) in the sewer system. Results of [5] also revealed that isotopic tracing ($\delta^{18}O$, $\delta^{2}H$) regarding analyzing groundwater, wastewater and rainwater is relevant in assessing water pathways on an urban catchment.

Besides stable isotopes, also the radioactive isotope of hydrogen, tritium (^{3}H) can be used for tracing sources and estimating of volume of extraneous water entering a sewer system. Tritium as a natural tracer is used to describe the timing and circulation patterns in hydrology and hydrogeology research, to estimate the natural groundwater recharge (eg. [20–22] besides, it is suitable for estimating mixing conditions by giving complementary information to understand and quantify freshwater contributions to the aquifer as well [18]. In regard of wastewater tritium as a tracer have been used for studying wastewater movement toward groundwater (i.e. aquifer vulnerability) or surface water, especially in case of nuclear power plants. [31] examined the spatiotemporal variability of a variety of tracers of wastewater and their movement to groundwater in fractured sedimentary bedrock aquifer in Ontario, Canada. Regarding tritium, they found that mixing was occurring between older groundwater and recently recharged water with elevated tritium level due to precipitation impacted by a nearby nuclear station.

The range of stable isotopes of drinking water (i.e. groundwater) and precipitation can overlap. In Hungary, stable oxygen isotope values of groundwater and precipitation range from −14 to −9‰ [16,23–25] and from −16 to +4‰ [15], respectively. Contrastingly, while tritium in precipitation is varying between 5 and 20 TU [26], tritium in drinking water is likely zero, since it is supposed to be exploited mainly from artesian waters having long residence times. In the large recharge areas in Hungary, the tritium concentrations are found to be zero below the ground level of 15–25 m [16,20].

The aim of this study is to assess the connection of increased flow rate and rainfall event in case of a separate sewer system in Tiszavasvári, Hungary, applying an isotopic tracer method, assuming that the tritium concentration of the drinking water is zero, and that stable isotope composition of the water supply and the local precipitation are different. Besides, the sensitivity of the applied methods and the extent of domestic connections of precipitation release that may lead to the abovementioned results in case of rain or storm events, are also to be estimated.

2. Materials and methods

2.1. Study area

The study was conducted in Tiszavasvári between July 22 and 27, 2017. Tiszavasvári is located in the northeastern Hungary. The geological subsurface of the area is composed of fine-grained Pleistocene sediments covered by loamy and clayey loess layers. The local climate is moderately warm and dry, with multi-decade (1971–2020) mean annual temperature of 10.5 °C and a mean annual precipitation of 540 mm [27].

The municipal water system is supplied with treated water from a sandy-loamy aquifer by five wells with an average flow rate of 1300 L min^{-1}, screened at 100–140 m depth. Average initial and working watertable of the wells are at 2.7 m and 11.5 m depth, respectively. The drinking water system delivers daily about 900 m^3 via 77.8 km of pipes. The sewer system has a length of 81.4 km and encompasses about 4000 residents resulting in roughly 330,000 m^3 yr^{-1} inflow. The sewer system is separated.

2.2. Sampling and analysis

Building upon the isotopic differences of precipitation and drinking water, we proposed a qualitative and quantitative analysis of rainwater assumed to be drained into the sewer system.

An ideal sampling run requires a short (maximum some hours long) rainfall event with longer dry periods before and after it. Thus, well-defined differences in isotopic composition of wastewater samples collected in the three stages can be expected. In the studied case rainfall started on July 24, 2017 at about 21:15 and lasted about 30 min. A continuous series of samples were taken from July 22, 2017 9:00 to July 27, 2017 21:00. In the dry periods, the sampling frequency was determined by taking into consideration daily changing of inflow rate of wastewater. Accordingly, samples for isotope measurements were taken twice a day, at 9:00 and 21:00, when maximum volume and average composition of wastewater can be observed. From the start of rainfall wastewater samples were taken in every 15 min until 23:00, in every hour after rainfall in the descending phase and later twice a day again. Flow rate data have been provided by the wastewater plant (Table S2).

Rainwater samples were collected in plastic vessels with diameter of 50 cm using a plastic funnel on three sites in the town. Groundwater from production wells and drinking/tap water were sampled into plastic bottles. Sampling sites are shown in Fig. 1.

Preparation of wastewater samples required multistage/step

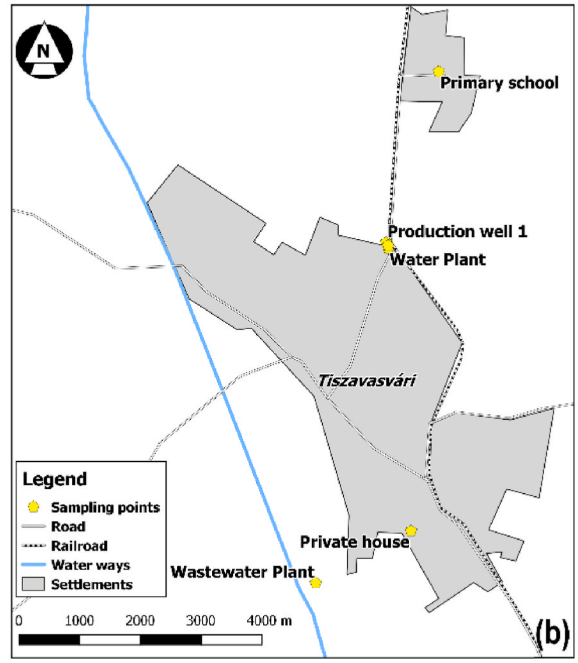

Fig. 1. Location of study area (a) and sampling sites (b) for wastewater (Wastewater Plant), rainwater (Primary school; Water Plant; Private house), drinking water (Water Plant) and groundwater (Production well 1,2).

filtration with varying pore size (20–0.45 μm) to remove solid particles of different sizes. Thereafter, samples were still considered biologically active, therefore, they were submitted to a pre-distillation that was followed by another distillation in the laboratory. For tritium analysis, the water sample was filled into a stainless steel vessel, and then the dissolved gases were pumped away from the water by a vacuum pump to eliminate the dissolved helium. After a few-week storage time, the ^{3}He content (as a daughter isotope of tritium) of the samples was determined by a Thermo Scientific HELIX SFT noble gas mass spectrometer, and then ^{3}H concentration was calculated from the ^{3}He of tritium decay [28,29]. Stable isotope analysis was carried out by a Los Gatos Research produced Triple Liquid Water Isotope Analyzer (LGR T-LWIA, model: 912–0050) based on laser absorption spectroscopy method. Flow rate is continuously monitored at the wastewater treatment plant and data were provided for this research. The precision of the measurements is better than ±2% above 1 TU (tritium unit) for tritium, ± 0.15‰ for δ^{18}O and ±1‰ for δ^{2}H.

3. Results and discussion

Municipal wastewater flows generally follow diurnal patterns that are largely controlled by actual production of domestic sewage. Fig. 2 presents diurnal variations of flow rate demonstrating the short-term fluctuation (measured in 15 min steps) under dry weather conditions and the effect of a rainfall (flow rate data can be found in Table S2). Flow rate curves of five out of the six observed days follow the same variation trends. Extraneous water flows, especially sudden inflows are generally detected based on the daily variation of wastewater flow rates.

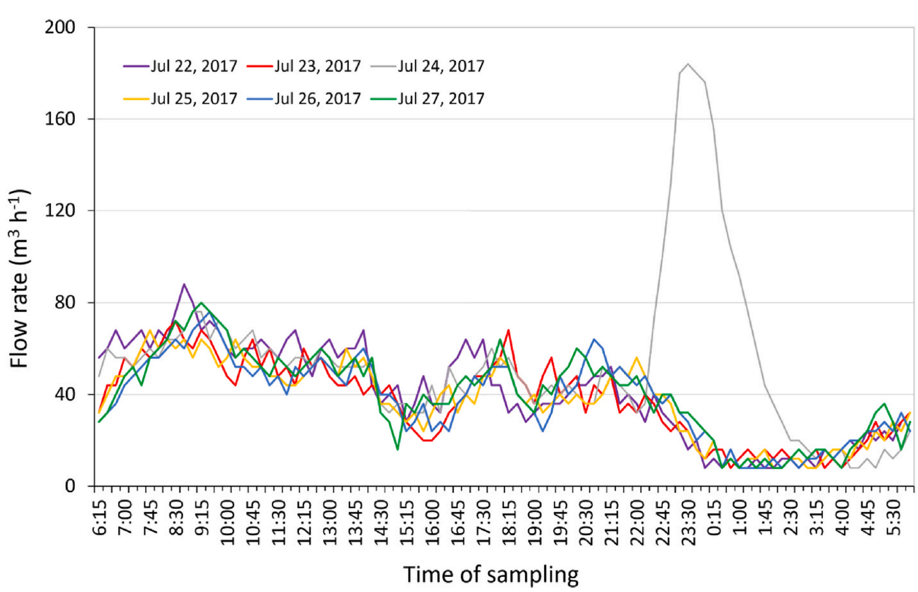

Fig. 2. Wastewater flow rate during the period of July 22 to 27, 2017.

Fluctuation of inflow in 15-min resolution during the six-day-long sampling can be seen, where typical daily maximums and minimums are observable. Under dry weather conditions the daily flow rate fluctuation is approximately 3.8 m^3h^{-1}, with a distinct difference between daytime and nighttime fluctuations. During daytime (6:00–24:00), there is a high fluctuation in the flow rate with the uncertainty of 4.4 $m^3 h^{-1}$. The nighttime (0:00–6:00), when wastewater flow is at the minimum level, fluctuation of the flow rate is lower (2.6 m^3h^{-1}) due to the significantly decreased water consumption of the inhabitants (daytime and nighttime intervals have been defined arbitrarily).

The rainfall event on July 24, 2017 is clearly noticeable, with a sudden increase of flow rate in the declining phase of daily discharge. This results in an increase of the flow rate from 30 to 40 m^3h^{-1} to 184 m^3h^{-1} in 75 min. The mean daily inflow of 934 m^3 during dry conditions rose to 1885 m^3 as a result of the 30 mm precipitation event (average of 32, 30 and 29 mm registered at the three rainwater sampling sites).

Mean values of tritium, $\delta^{18}O$ and δ^2H of wastewater in the dry period prior to the rainfall event (0.37 ± 0.06 TU, −11.38 ± 0.16‰ and −81.50 ± 0.66‰, respectively) largely reflect the composition of the tap water (0.13 ± 0.01 TU, −11.90 ± 0.10‰ and −85.63 ± 0.21‰, respectively) (Table 1) as the dominant source of wastewater. The small difference may be attributed to organically bound isotopes from plant or animal food products or shallow groundwater infiltration due to possible system failures.

The $\delta^{18}O$ and δ^2H values of the groundwater (−11.93 ± 0.16‰ and −84.89 ± 0.18‰, respectively) reflect recharge in cold climate conditions. The water here is exploited from 100 to 140 m depth where the groundwater is of late Pleistocene age [23]. The isotope composition of the water from the precipitation event is significantly different from that of the groundwater, allowing to easily separate the rainwater from the wastewater.

Fig. 3 shows how tritium, $\delta^{18}O$ and δ^2H values as well as flow rate changed during the rain event. Soon after it had started raining, there was a rapid increase in discharge closely followed by the change of isotopic values reaching their maximum values (184 m^3h^{-1}, 8.63 ± 0.11 TU, −7.32 ± 0.07‰ and −46.02 ± 0.39‰ for flow rate, tritium, $\delta^{18}O$ and δ^2H, respectively). This sudden increase of discharge was followed by a rapid fall until flow rate reached its usual nighttime minimum values (around 10–15 m^3h^{-1}). Isotopic composition reached the starting values in a more moderate pace suggesting that in spite of rapid falling of flow rate, some proportion of rainwater in the total wastewater is still notable. At 9:00 on 25 July another peak can be seen in the flow rate and isotope values, indicating that increased discharge deriving from morning residential water consumption flushed residual precipitation out by speeding its flow up.

There is an obvious strong correlation between tritium and stable isotope data ($R^2_{\delta 18O}$ = 0.9552; $R^2_{\delta 2H}$ = 0.9743), therefore both are suitable for tracing extraneous waters and estimating quantity of them. However, sensitivity of the tracing method may vary depending on the applied isotopic tracer.

Changes in isotopic composition of wastewater should thus reflect the varying proportions of foul sewage and rainwater contributing to the total wastewater discharge. An overview of the $\delta^{18}O$ and δ^2H values of all samples collected during our experiment are given in Fig. 4. It gives an overview of $\delta^{18}O$ and δ^2H values of all samples collected during our experiment. The stable isotope composition of the mixing end-members is sufficiently distinct, with mean rainwater values plotting at $\delta^{18}O$ = −5.25 ± 0.17‰, δ^2H = −30.90 ± 1.03‰ and groundwater and drinking water values plotting at $\delta^{18}O$ = −11.93 ± 0.16‰, δ^2H = −84.89 ± 0.18‰ and $\delta^{18}O$ = −11.90 ± 0.10‰, δ^2H = −85.63 ± 0.21‰, respectively. Binary mixing of wastewater and rainwater is obvious, with the isotopic composition of the wastewater varying within the range covered by the two mixing end-members. During dry periods, the stable isotope values of wastewater (sourced predominantly from drinking water) are close to the drinking water values, as expected. Due to the rainfall dilution of wastewater, isotopic composition of wastewater samples collected during and right after the rainfall event are found on the half way between that of drinking water and the rainwater.

Based on the stable isotope methodology of [4], in this study the amount of precipitation inflow to the sewer system was estimated using the general assumption of binary mixing. Integration of rainwater discharge over the rain event and the following hours, gives a total discharge of parasitic inflow of 1885 m^3. Taking account of the amount of fallen precipitation (30 mm), rainwater fallen onto approximately 57000 m^2 has drained into the sewer system. This is about 1% of the populated area. If one assumes that this rainwater is led into the sewer system directly from the roof with a size of about 100 m^2, then 570 establishments can be estimated to have illegal connections to the sewer system. Of course, there is no evidence that extraneous rainwater comes from the roof area.

According to the findings of the recent study, all three isotopic tracers applied in this research are suitable to trace inflowing parasitic precipitation, however, differences in the sensitivity can be observed. Comparing the methods applied for detecting precipitation in wastewater, it can be concluded that under dry weather conditions isotopic tracers have nearly constant or slightly varying values, independently from the daily fluctuation of wastewater flow rate. Daily flow rate of wastewater changes in a range of 2–80 $m^3 h^{-1}$ resulting from the varying rate of communal water consumption throughout a day. Additionally to the daily variation of the discharge, it has a short-term fluctuation (measured in 15-min steps). Due to this variation, detecting a smaller amount of precipitation flowing into the sewer by applying the flow rate method seems to be less accurate than in case of isotope tracers, since increase in flow rate caused by a moderate rainfall event may be obscured by diurnal fluctuation. Instead of daily mean flow rate, variation of discharge (difference of consecutive flow rate values) is considered more reasonable to track increase of discharge caused by a rainfall event.

In case of 95 and 99.7% confidence range (2σ and 3σ uncertainty) variation of discharge still may be on the edge or out of the range. As it is presented in Table 2, daytime minimum detectable excess (MDE, defined as two or three times the standard deviation) of discharge variation is 18.9 m^3h^{-1}, though, the variation may reach (or slightly exceed) 20 m^3h^{-1} in dry periods increasing the uncertainty of detection of small amount of parasitic rainwater. Similarly, nighttime variation of discharge may collide with MDE (3σ) (12.2 m^3h^{-1}).

However, applying isotopic tracers, even a few percent increase in discharge is detectable due to the favorably large isotopic difference of the end-members. Tritium proved to be the most sensitive in this respect, allowing as low as about 2% (0.62 m^3h^{-1}) of parasitic precipitation to be traced. Similarly, $\delta^{18}O$ and δ^2H are suitable for detecting a small amount of rain (8% and 4%, respectively) compared to the total discharge (69.95% at nighttime and 28.64% at daytime). These isotope methods

Table 1
Isotopic composition of groundwater (GW), drinking water (DW), wastewater (WW) (under dry weather conditions) and rainwater (RW).

Date of sampling	Sample	Tritium (TU)	$\delta^{18}O$ (‰, VSMOW[a])	δ^2H (‰, VSMOW)
24/7/2014	GW	0.07 ± 0.01	−11.93 ± 0.16	−84.89 ± 0.18
20/7/2014	DW	0.13 ± 0.01	−11.90 ± 0.10	−85.63 ± 0.21
22/7/2014, 9:00	WW1	0.37 ± 0.02	−11.36 ± 0.07	−81.59 ± 0.32
22/7/2014, 21:00	WW2	0.32 ± 0.01	−11.19 ± 0.04	−81.03 ± 0.30
23/7/2014, 9:00	WW3	0.45 ± 0.02	−11.40 ± 0.09	−80.09 ± 0.32
24/7/2014, 9:30	WW4	0.34 ± 0.02	−11.57 ± 0.05	−82.40 ± 0.27
24/7/2014	RW1	8.97 ± 0.17	−5.31 ± 0.07	−30.38 ± 0.49
24/7/2014	RW2	9.13 ± 0.18	−5.38 ± 0.07	−32.09 ± 0.29
24/7/2014	RW3	9.24 ± 0.18	−5.06 ± 0.09	−30.24 ± 0.30

[a] Vienna Standard Mean Ocean Water.

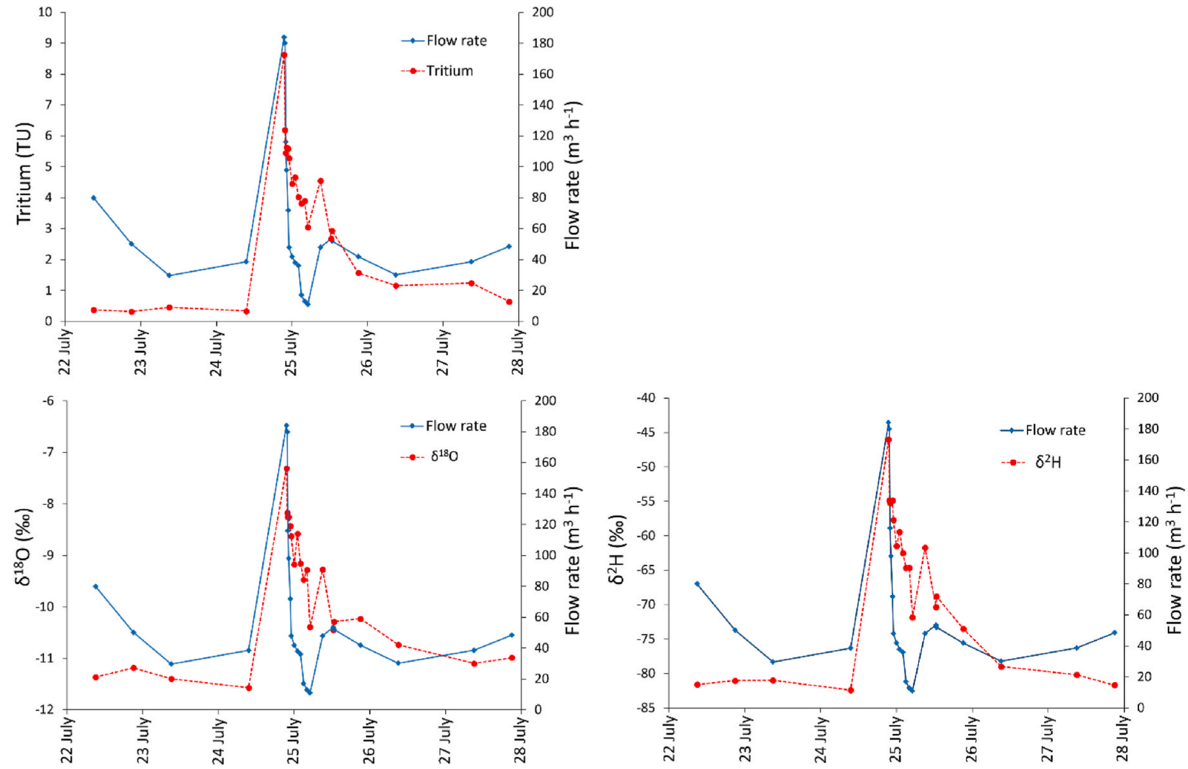

Fig. 3. Comparison of flow rate and water isotope values from 6:00 on July 22, 2017 to 24:00 on July 28, 2017.

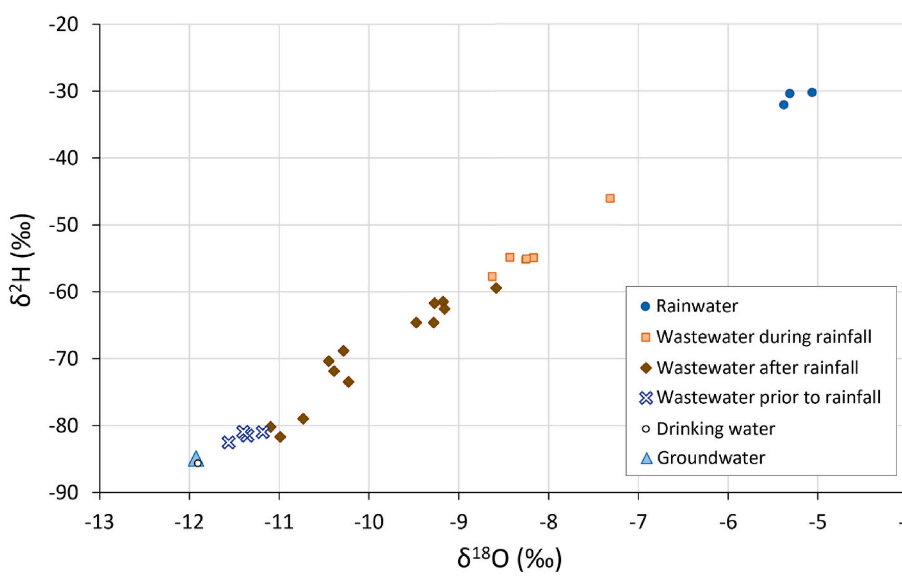

Fig. 4. Isotopic characterization of groundwater, drinking water, wastewater and rainwater.

are far more sensitive than the flow rate method.

4. Conclusions

This study confirmed, through the example of a small town (Tiszavasvári) in Hungary, that isotopic tracing is a suitable method for detecting parasitic rainwater in a separate sewer system. During the observed rainfall event both tritium and stable isotope values (δ^2H, $\delta^{18}O$) of water changed in the same way as the flow rate. In addition, based on the δ^2H and $\delta^{18}O$ data, a binary mixing of wastewater and rainwater has been verified. According to the dataset of this study, the applied isotopic tracers are more sensitive tools for detecting and quantifying the rainwater joining the sewer system than using solely the discharge method. The detectable proportion of precipitation is orders of magnitude less in case of isotopic tracers than in case of discharge variation. However, it has to be noted that the seasonality of the isotopic composition of precipitation has a major impact on the applicability of the method. Aiming for larger isotopic difference of the end-members, in case of groundwater recharged under cold climate conditions (i.e., resulting in lower δ values), a summer sampling experiment is ideal, while in the other case sampling in the winter half-year is suggested. Considering the annual tritium trend of precipitation, if tritium is the applied isotope for tracing, it is suggested to sample during summer when tritium content of rainwater is higher than in the winter half-year.

Table 2
Minimum detectable excess (MDE) of the applied methods and the adherent estimated amount of precipitation mixed into wastewater. (MDE = dry weather mean + 2 or 3σ).

	Tritium (TU)	$\delta^{18}O$ (‰, VSMOW)	$\delta^{2}H$ (‰, VSMOW)	Variation of discharge[a] ($m^3\ h^{-1}$)	
				nighttime	daytime
Dry weather mean (1σ)	0.37 ± 0.06	−11.38 ± 0.16	−81.50 ± 0.66	4.22 ± 2.65	6.42 ± 4.16
Minimum detectable excess (MDE) (2σ)	0.49	−11.06	−80.18	9.51	14.74
Minimum detectable excess (MDE) (3σ)	0.55	−10.91	−79.52	12.15	18.91
Estimated proportion of rainwater at MDE (3σ), %	2	8	4	69.95	28.64
Estimated rainwater discharge at MDE (3σ), m^3h^{-1}	0.62	2.49	1.25	12.15	18.91

[a] Difference of consecutive flow rate values.

The ideal sampling run should be carried out after a longer dry period to make sure that possible residual rainwater of the previous precipitation event has fully disappeared. Avoiding larger fluctuation of flow rate, nighttime sampling is suggested. Though, tritium method proved to be the most sensitive to trace extraneous water, cost and complicacy of the method also has to be taken into consideration. Stable isotopes also have notable sensitivities and more favorable costs. Water isotope values were also found appropriate to evaluate the quantitative contributions of precipitation inflowing to the system and to estimate the number of households that discharge rainwater to the sewer system.

Credit author statement

Anita Puskás-Preszner: Data curation, Investigation, Formal analysis, Writing – original draft preparation, **Anna Orosz**: Investigation, **László Kompár**: Investigation, Conceptualization, Methodology, **Marianna Túri**: Investigation, Visualization, **László Palcsu**: Supervision, Writing- Reviewing and Editing.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Laszlo Palcsu reports financial support was provided by European Regional Development Fund.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.rineng.2022.100360.

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