Highlights

Better understanding in-sewer processes with on-site continuous mass spectrometric analysis of sewer gases

Mengqi Zhu, Matthias S. Brennwald, Jes Vollertsen, Rolf Kipfer, Jörg Rieckermann

- For the first time, a portable mass spectrometer has been used to measure noble and other gases continuously in sewers for several weeks
- Simultaneous monitoring of multiple gases can provide novel insight into sewer transport and transformation processes
- The 'tracer system' of noble gases and portable mass spectrometry is promising to better understanding sewer-groundwater interactions
- Open issues concern robust sampling techniques and the impact of boundary conditions, such as sewer ventilation

Better understanding in-sewer processes with on-site continuous mass spectrometric analysis of sewer gases

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Abstract

Sewer systems are an important part of the urban water cycle, because they protect public health and natural aquatic environments from contamination. However, adapting our sewer systems to future challenges requires a better understanding of important transport and transformation processes, such as emission of greenhouse gases or unwanted infiltration of clean groundwater. In this short communication, we propose that monitoring of gases with mass spectrometric methods can deliver a better insight into biochemical transformation and exchange processes than what is currently possible with conventional wastewater analytics. The results from three case studies provide insight into the dynamics of GHG, such as CH_4 and CO_2, and use Helium as a tracer to assess both groundwater infiltration and sewer airflow patterns. In the future, this can be used to design novel experiments, e.g. by simultaneously analyzing inert and active gas species, or actively injecting tracer gases into the ground- and wastewater. Future work should investigate better filtration or indirect sampling methods to sample gas from sewage, such as specially-tailored flux chambers. For network-wide analyses, boundary conditions regarding sewer ventilation, as well as sources and sinks of gases need to be better understood.

Keywords:

noble gases, sewer, portable mass spectrometer, tracer system, wastewater pollution, infiltration, sewer gas exchange, sewer transport and transformation

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1. Introduction

Sewers are important because they protect public health and natural aquatic environments from contamination by safely conveying polluted wastewater to treatment plants. Although sewers are expected to last about 75 to 150 years, it is often much less in reality as current maintenance is insufficient given reduced public expenditure. The resulting sewer deterioration causes problems such as sewer collapse, blockages from infamous “fatbergs”, sulfide corrosion, odor problems, and unwanted intrusion of clean ground- and stormwater, so-called “infiltration and inflow” (Ellis et al., 2010; Staufer et al., 2012; Vollertsen et al., 2008).

With rapid urbanization and climate change, these problems will become worse. Hence there is an urgent need to adapt the sewer systems that were designed and built by previous generations, to a future with more diverse needs than just hydraulic capacity. As an example, the integrated assessment of sewers and wastewater treatment plants is no longer only based on flows and pollutant loads, but already takes into account Greenhouse Gas (GHG) emissions and the potential to recover energy and other resources from the wastewater system (Figueroa et al., 2021; Yadav et al., 2021; Saagi et al., 2022; Di Capua et al., 2022). This requires a more detailed understanding of transport and transformation processes of the compounds in the sewers than before.

Transport and transformation processes in sewers are usually highly dynamic, especially during wet weather, and pollutant concentrations often change within a few minutes (Huisman et al., 2004; Rieckermann et al., 2005). Unfortunately, conventional sewer measurements of water levels and velocities only provide limited insights on transformation processes. Therefore, tracer systems, which we define as the combination of tracer substance and monitoring system, are increasingly used to study in-sewer processes. For example, natural tracers such as sulfate (SO_4), chemical oxygen demand (COD), the artificial sweetener acesulfame, caffeine, and stable isotopes of water were used to separate the hydrographs of combined sewage into groundwater (stormwater) and foul sewage (Heiderscheidt et al., 2022; Houhou et al., 2010; Kracht et al., 2008; Zhao et al., 2020). In a similar fashion, gaseous tracers such as sulfur hexafluoride (SF_6), carbon monoxide (CO) or oxygen (O_2) were utilized to study the horizontal gas transport and ventilation in sewers (Huisman et al., 2004; Madsen et al., 2006; Parker and Ryan, 2001). However, most of the current tracer systems rely on grab sampling followed by a labor-intensive and time-consuming lab-based analysis. Moreover, tracer systems which can be used to observe in-sewer processes with the required temporal resolution of a few minutes are largely lacking.

In this short communication, we present a new set of tracer systems to study sewer transport and transformation processes with a portable mass spectrometer. We used it to perform the very first continuous and simultaneous high-resolution on-site measurements of noble gases and greenhouse gases in sewers and demonstrate the potential in three case studies: i) online monitoring of biochemical gases to assess dynamics and GHG emissions, ii) using noble gases to monitor groundwater-sewer interactions and iii) monitor sewer airflow patterns. In addition, we critically discuss opportunities to use portable gas monitoring to better understand in-sewer processes, identify current limitations and suggest future research to overcome them.

2. Materials and Methods

2.1. Online measurements of gases in sewers with a portable mass spectrometer

We used a portable mass spectrometer (miniRUEDI, Gasometrix Switzerland; Brennwald et al. (2016)) to quantify the partial pressures of the gas tracers: helium (He), argon (Ar), krypton (Kr), carbon dioxide (CO_2), methane (CH_4), in the sewer headspace. The 6-port inlet of miniRUEDI is capable of continuously sampling up to six gas sources alternately. In the following case studies, the port selector switched between the sewer atmosphere and standard gases. The signals of the gases in the sewer headspace were calibrated by comparing peak heights with the standards and then converted to partial pressures and volumetric percentages (ppm). For He, Ar, and Kr, atmospheric air was used as a standard for the calibration, whereas for CO_2 and CH_4 a standard gas mixture of 1% CO_2 and 1% CH_4 in N_2 was used (Brennwald et al., 2016; Mächler et al., 2012).

In this study, the gases were sampled from the headspace of the sewer system with PVC tubes (ϕ 6 mm or 8 mm, Festo) and peristaltic pumps (PD 5001, Heidolph). The gases from the headspace were pumped to the miniRUEDI, which was located at the ground level. The purging time of the sampling systems depends on the length of the tubes, ranging from 1 to 2 min in this study. The overall temporal resolution for the analysis of one sample (from one single port) was around 5 to 8 minutes including the purging time before switching the port. In comparison to grab sampling, even with automatic samplers, this makes it possible to better capture the dynamics of the gas tracers and associated processes.

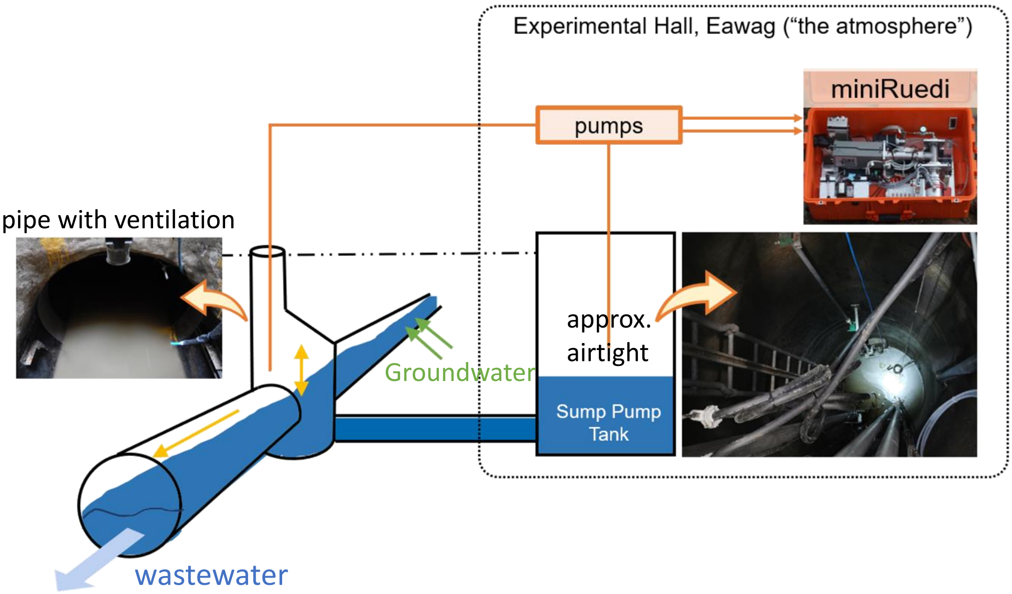

Figure 1: Scheme of miniRUEDI online measurements. The portable mass spectrometer miniRUEDI was fixed in a 80 cm × 52 cm × 32 cm hard-shell suitcase with wheels. The overall weight was around 32 kg. The gases from the sewer pipe headspace and the headspace of the sump pump tank were sampled with the PVC tubing and peristaltic pumps. Their signals were then analyzed by the miniRUEDI.

2.2. Study site

The study catchment is located in the municipality of Dübendorf, close to the Swiss Federal Institute of Aquatic Science and Technology (Eawag). Dübendorf consists of a mixture of infrastructure without a dominant type of industry. The wastewater composition is mainly residential, with some industrial contributions. Experiments were conducted in one of the gravity-driven trunk sewers that collects and conveys the combined wastewater from the town through the domain of Eawag. It has a circular profile with a diameter of 1000 mm, an average depth around 2.8 m, and a slope of 1.6‰. Long-term monitoring data from an area-velocity meter (OCM pro CF, NIVUS, Germany) showed average dry weather flow rates between 50 L/s and 115 L/s. The groundwater infiltration flow rate, estimated by the night-time minimum method (De Bénédittis and Bertrand-Krajewski, 2005), is about 45 to 50 L/s. Although the nigh-time minimum methods makes simplifying assumptions and neglects dynamics (Kracht et al., 2007), the range is typical for combined sewer systems.

In our study, we sampled this location at two points, one with natural ventilation and one without (Figure 1). The main monitoring point is inside a manhole of the trunk sewer (47.404770, 8.611741). This was readily accessible as an underground cable canal connecting the manhole to the experimental hall of Eawag, where most of our wastewater research is performed. A second monitoring point was located inside the tank of a sump pump station (labeled as "pstank"), which is used to continuously extract wastewater from the trunk sewer to the experimental hall. The tank is 6 m deep, with a residence time of ca. 30 min. In contrast to the trunk sewer, it is virtually air tight. Thus, it can be assumed that in the sump pump tank the equilibrium state of the studied gases between bulk wastewater and corresponding headspace has been established. The concentration of the gases dissolved in the bulk wastewater can therefore be calculated by Henry's law and the partial pressures of the gases in the headspace.

In addition to the gas measurements and the flow data in the sewer, we collected complementary monitoring data on precipitation, air pressure, air temperature, etc. at the study site from the official MeteoSwiss weather station at Zurich-Kloten (47.478387, 8.530160, ca. 10 km away) and from the station "Dübendorf-Empa" of the Swiss National Air Pollution Monitoring Network (NABEL) (47.40299, 8.61341, ca. 0.5 km away). These data sets are important as reference measurements to calibrate the miniRUEDI to local ambient air and to interpret the results. Further complementary data have been collected, such as atmospheric gases concentrations (case study 1 and 2) and dissolved noble gases concentrations in the local groundwater (case study 2). The raw and processed data are openly available at the The Eawag Research Data Institutional collection (ERIC [1]) for further analysis.

3. Results and discussion

In the following, we present the results from three case studies, which demonstrate the usefulness of gas tracers and mass spectrometric analysis in studying in-sewer processes. Case study 1 investigated the gases related to biochemical reactions in the sewers. Case study 2 used helium as a groundwater tracer to assess groundwater infiltration, arguably making rather rough assumptions. In case study 3, we artificially injected helium as a tracer to measure the air transport and ventilation in a short reach of the trunk sewer.

3.1. Case 1: online monitoring of biochemical gases

CO_2 and CH_4 are key gas species related to biochemical reactions and GHG emission. They were hence selected as gas tracers, and their concentrations in the pipe headspace and the sump pump tank headspace were monitored and quantified as described in section 2.1. Each sampling point was connected to one inlet port of the miniRUEDI. The device measured the gases from these two points semi-simultaneously by continuously switching between the two inlet ports every 6 to 7 min.

An example of online monitoring results of gas concentrations are shown in Figure 2. The CO_2 concentration in the sump pump headspace was about 2000 ppm, while the CH_4 concentration was about 750 ppm. The concentrations of these two gases in the sewer pipe headspace were about a third to a fourth lower (750 ppm and 200 ppm, respectively), most probably due to ventilation losses and dilution. The measured CH_4 concentrations were close to the lowest bound of the values reported in other studies (Liu et al., 2015a,b). However, due to the nature of our study, this data may not be representative for the whole study site, as the monitoring campaign was not designed to capture the seasonality or spatial distribution in the catchment.

[1]Please see: https://doi.org/10.25678/0006JS.

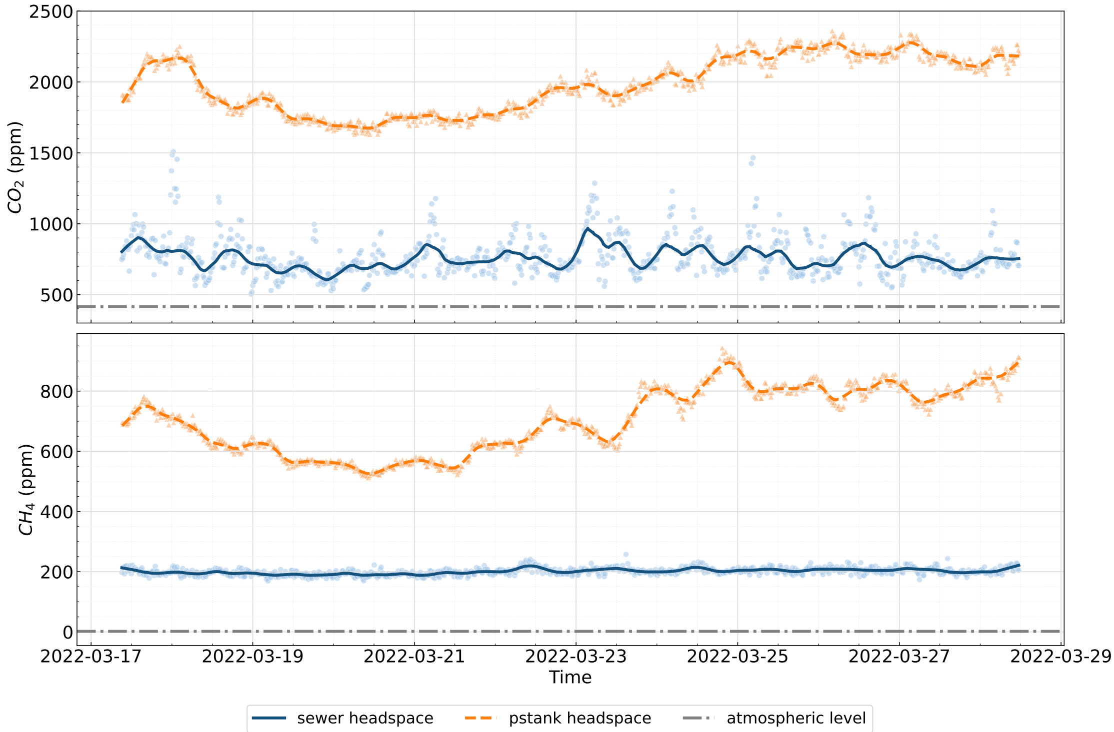

Figure 2: shows the concentrations of CO_2 and CH_4 in the ambient atmosphere, in the pipe headspace (connected to the ambient atmosphere) and the headspace of the sump pump tank (not ventilated). The open circles show the original data points while the lines show the results of a LOESS smoothing. The atmospheric concentrations were derived from typical fractions of the gas species in the atmospheric air. It can be seen that the concentrations in the headspace of the sump pump tank were higher than those in the pipe headspace, because the gases produced by biological activities were accumulated. The CH_4 concentrations are in the lower range compared to literature values (Liu et al., 2015a,b)

The online measurement of CO_2 and CH_4 with high temporal resolution better captures the dynamics of the gases compared to traditional grab sampling and offline analysis. Such datasets enable various advanced data analysis when coupled with conventional flow measurement. For example, an exploratory data analysis using these datasets and linear regression suggested a slightly positive correlation between the sewer temperature and the CO_2 concentrations in the pipe headspace (0.0896, normalized). The novel data and resulting analysis therefore provide new insights into relevant processes, such as pollutant loading and GHG emissions. Thus, they can play a role in assessing mitigation strategies. However, up till now only the information regarding gas concentrations can be acquired. Gas flux, as a more critical indicator in many sewer system studies, can only be accessed if more information and knowledge about sewer air transport and ventilation is available (see section 3.4).

3.2. Case 2: continuous monitoring of noble gases to assess groundwater infiltration

In the second case study, we measured noble gases in the sewer pipe headspace and sump pump tank headspace for the first time. Because groundwater is usually enriched with noble gases above atmospheric solubility equilibrium (Kipfer et al., 2002),the noble gases can be used to estimate the magnitude and the dynamics of infiltration into sewers. Generally, the sampling and the gas analysis procedure are the same as shown in section 2.1.

A prior investigation showed that the concentrations of the dissolved noble gases in the local groundwater were higher than those in the atmosphere-saturated water by up to 20% (Popp et al., 2020).Without groundwater infiltration, the headspace concentrations of noble gases in the "pipe" would be the same as atmospheric values. However, the observed concentrations in the monitoring points were slightly above the atmospheric concentrations. Although the observed average increase of 0.73% was small compared to the analytical precision of the miniRUEDI (<5%), this might still indicate the presence of groundwater, as it is the main source of noble gases in the sewers apart from atmospheric air. However, the observed dynamics of the noble gases were not synchronized with the sewer flow rate (see below).

Suggested by Kracht et al. (2007), a simplified mass balance equation of infiltration ratio based on end-member mixing analysis was used to estimate the ratio of groundwater infiltration, which was converted into absolute values by multiplying with the flow data. For the sake of the argument, since direct liquid-phase sampling methods are lacking, we made the rather rough assumption that equilibrium was established between the foul sewage from the households and the atmospheric air. We also assumed that the bulk wastewater in the sump pump tank and the corresponding headspace were in equilibrium, given the virtually airtight conditions of the well. The dissolved noble gases in the foul sewage and the bulk wastewater were then derived based on Henry's law using atmospheric data and on-site measurement data from the miniRUEDI. Another simplifying assumption was that

10% of the noble gases input was lost from the sewer system due to ventilation. The details on the assumptions and calculations can be found in Zhu (2022).

The concentration profiles of dissolved helium, as an example of noble gases, and the groundwater infiltration flow rate were calculated as shown in Figure 3. In general, the estimated infiltration flow rate ranged from 25 L/s to 80 L/s (infiltration ratio 40% to 80%), which is similar to the estimate obtained with the night-time minimum method (Figure 3). In comparison, values from literature and previous research on other catchments range from 15% to 80% infiltration, making our values close to the higher bound (Kracht et al., 2007; Zhao et al., 2020). Interestingly, the daily fluctuations suggested by our observations corresponded to the observations using stable isotopes of water as tracers and a fast grab sampling method (Kracht et al., 2007).

In general, the on-site continuous monitoring of noble gases as tracers allowed exploring the groundwater infiltration dynamics down to the minute scale. Such measurements could give unprecedented insights into the short-term dynamics of groundwater-wastewater interactions and greatly improve asset management strategies. It should be noted that, due to the lack of direct liquid-phase sampling methods, the mass balance relied on rough assumptions regarding ventilation conditions and gas exchange in the sewers, which could lead to a systematic bias. Further, the spatial distribution of groundwater infiltration in the drainage catchment was not considered. The routing of infiltrated groundwater could have a great impact on the dynamics and kinetics of gases in the sewer and hence affect the quantification of groundwater infiltration even more.

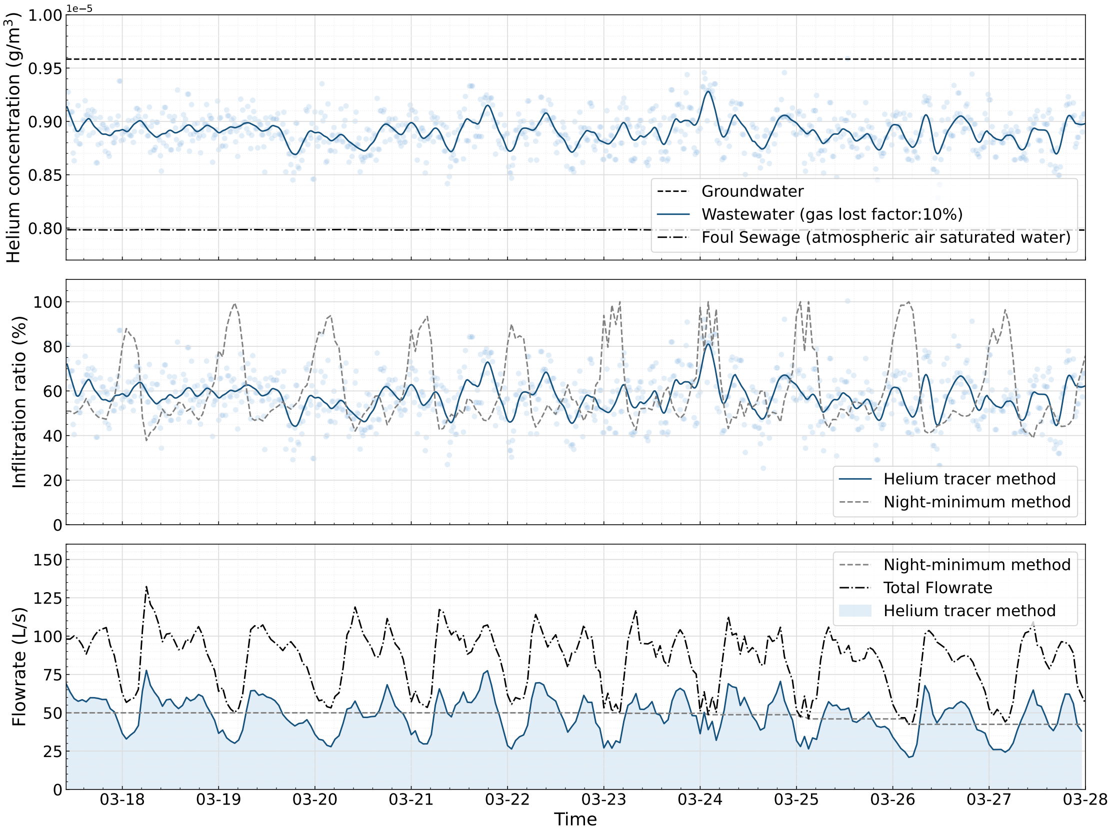

Figure 3: (Top) Time series of helium concentrations in the wastewater, groundwater and the foul sewage at the study site, (middle) the temporal dynamics of the estimated infiltration ratio, and (bottom) the resulting hydrograph separation. In the upper figure, the open circles describe the raw monitoring data in the sump pump tank, while the blue line shows the smoothed results of LOESS. The foul sewage was assumed to be in equilibrium with the atmospheric air. The dotted line showed the dissolved helium in the local groundwater. The line located in-between the two end-members suggests the existence of groundwater in the wastewater. In the middle and bottom plots, the results of infiltration ratio and associated hydrograph decomposition suggest that the estimates of the helium tracer method are in a similar range as those computed with the conventional night-time minimum method. Although the absolute infiltration values are based on rough assumptions, the observed daily fluctuation of mixing ratios are rather robust.

3.3. Case 3: estimation of sewer air transport using helium as tracer

We measured the sewer headspace air velocity from a travel time experiment with helium. Specifically, we injected 9 L as a dirac input at a manhole 170 m upstream of the pipe monitoring location, assuming complete mixing over the entire headspace cross section. At the pipe, the helium signals were sampled and recorded as described in section 2.1. For a simplicity estimation of the travel time, we took the time difference between the injection and the first concentration peak (Figure 4) instead of fitting an advection-dispersion transport model. The resulting air velocity was 0.064 m/s and the flow rate 39.6 L/s, with a ratio of air velocity to water velocity of 9.1%. Both the air velocity and the velocity ratio are comparably low (Madsen et al., 2006; Parker and Ryan, 2001).

Interestingly, the observations did not show a smooth breakthrough curve, suggesting that air flow conditions were complex due to various structures. However, the results clearly demonstrate the potential for further research regarding sewer ventilation and air transport. For example, given a sewer reach with complete geometry information, one can use the mentioned tracer systems to quantify ventilation rates (i.e., the turnover of sewer air) via mass balances.

In practice, we found it challenging to dose a large volume of tracer gas as a dirac pulse from the surface through a closed manhole cover. Our final solution was to use a large balloon filled with Helium, which was attached to the tube of an air compressor with a nail placed inside. The compressed air pushed out the nail and popped the balloon, creating an instantaneous tracer pulse.

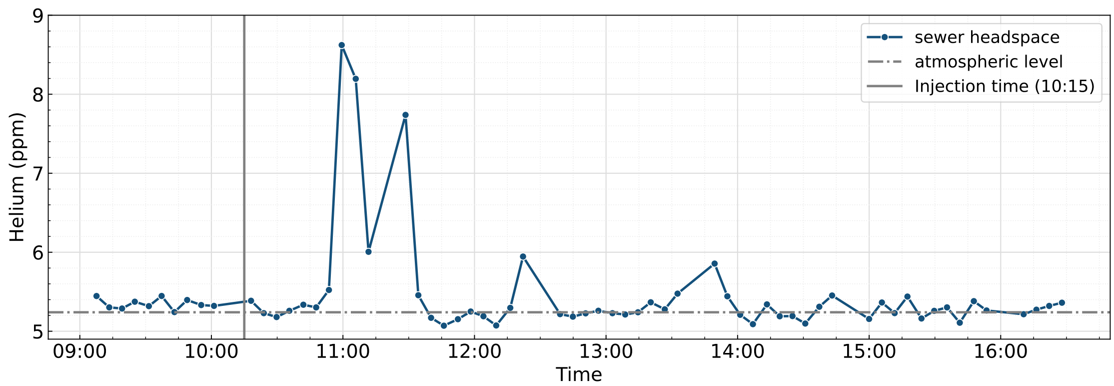

Figure 4: Time series of the helium tracer in the pipe headspace after a dirac input at time 10:15, in a manhole 170 m upstream. The first peak showed at around 11:00. The following small peaks might be caused by effects such as back-flow introduced by the various structures in the sewer. From this experiment, the average sewer air transport velocity was estimated to 0.064 m/s.

3.4. Opportunities and Challenges

Continuous monitoring of gases in sewers using portable mass spectrometers, like the miniRUEDI, was investigated in three case studies. These studies demonstrated its potentials for advancing research in various areas, including biological transformations, pollutant dynamics, groundwater infiltration, and sewer ventilation.

In our view, one of the biggest potential benefits is to measure several gases simultaneously with one single portable device like miniRUEDI. Combining information on biochemically relevant gases and inert gas tracers could possibly construct new approaches for non-contact based pollution monitoring. Also, together with pumps for autonomous dosing of multiple tracers, the in-situ measurements of gas tracers at a high temporal resolution allows us to assess important sewer process parameters relevant for sewer modeling and support subsequent maintenance decisions. For instance, by injecting multiple inert tracers with different Henry's constants, their travel time and concentrations can serve as valuable inputs for calibration of sewer air transport models, which is helpful for predicting odor emission, corrosion, and heat transfer. Such methodology could also be used to design more complex experiments, similar to the QUEST-C tracer method (Rieckermann et al., 2007). which dosed two distinct tracers at different locations to assess wastewater leakage between the locations. In addition, in-situ measurements of gas tracers would also facilitate novel active tracer techniques. For example, it might become possible to inject noble gases into the groundwater and monitor them in the nearby sewer networks to assess the spatial variability of sewer-groundwater interactions.

However, as with every novel method, there are still substantial limitations. Firstly, we experienced that it is still challenging to sample gases directly from the liquid wastewater, which has much more particles than, for example, grondwater. The gas-equilibrium membrane inlet system developed for clean ground and surface waters (Mächler et al., 2012) would rapidly clog when applied in wastewater. For the proposed applications, only sampling the sewer atmosphere means that the mass balance of the tracers is always incomplete and that it can only be closed applying several rough assumptions on gas exchange and ventilation patterns. We see two possible solutions to this: first, a gas-tight clogging-free pre-filtration system coupled with a gas-equilibrium membrane inlet module could be developed. This probably requires some automation, including back-washing with filtrate to operate reliably under changing conditions. Second, one could develop a floating flux chamber as in Gruber et al. (2020) and Attermeyer et al. (2021), to facilitate the development of an equilibrium state.

Moreover, unknown boundary conditions, such as ventilation conditions and gas exchange with the atmosphere, make it difficult to interpret measurement data, and to extend the findings to the assessment of an entire drainage system with respects to topics like large-scale GHG emissions. We hope that more experience with mass-spectrometric gas monitoring will help to close this gap.

4. Conclusions

Adapting our sewer systems to future challenges from urbanization and climate change requires better experimental methods to improve understanding of transport and transformations in sewers. The results of our study suggest that the continuous monitoring of gases with mass spectrometric methods can deliver better insights into in-sewer processes than what is possible with current wastewater analytics. Specifically, online monitoring of gases related to bio-chemical transformations can provide insights into pollutants and their conversion processes. The noble gases also seem very promising as environmental tracers for sewer-groundwater interactions.

In our view, the most promising direction is probably the simultaneous analysis of inert and active gas species, which might provide new insights into the biological activities such as oxygen turnover in the sewers. In the future, this might give new insights into pollutant dynamics. Also, the possibility of using multiple noble gases simultaneously as artificial tracers, has not yet been explored in wastewater systems. Currently, one of the strongest limitations is that a gas-tight clogging-free pre-filtration system coupled with a gas-equilibrium membrane inlet module to monitor dissolved gases is missing. Also, it is challenging to interpret the data regarding system-wide processes, such as groundwater infiltration, which still requires rather rough assumptions. Therefore, future research should concentrate on better understanding the unknown boundary conditions regarding sewer ventilation and sources and sinks of gases.

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