AUTHOR'S REVIEW OF THE DOCTORAL (PHD) DISSERTATION

NATIONAL UNIVERSITY OF PUBLIC SERVICE
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Risk assessment of organic micropollutants in riverbank filtration based drinking water supply

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THE DETAILS OF THE SCIENTIFIC PROBLEM

The legal base for the quality requirements for drinking water is provided by Government Decree 201/2001 (X.25.) on the quality requirements for drinking water and the monitoring procedure. This Regulation is based on and fully complies with Council Directive 98/83/EC on the quality of water intended for human consumption. This Directive was later replaced by EU Directive 2020/2184. These set out the expectations and requirements for the quality of drinking water. The limit values set for certain pollutants are the base for producing healthy and safe drinking water.

The problem is, however, that a significant proportion of organic micropollutants are not included in drinking water quality regulations and routine water quality testing, because of the rather limited information available. There are exceptions to this, with some organic micropollutants being required to be tested in drinking water under the relevant regulation.

It is only in the recent decades that the scientific community has turned to research on these pollutants, and the results are still being published. One of the main problems is that, although our knowledge of the presence of organic micropollutants in the environment is constantly increasing thanks to the growing databases, we still lack of accurate information on their health effects. In many cases, it takes years or decades to know the health and ecotoxicological effects of a chemical. Partly due to this lack of information, the slow legislative system is only able to incorporate organic micropollutants into the applicable regulations and rules in a slow and long process.

To get closer to this goal, it is important to have a comprehensive and accurate view of the presence and amount of organic micropollutants in our drinking water aquifers. Although limited in quantity, there are now useful data series available that provide information specifically on the occurrence of organic micropollutants in our surface and groundwater aquifers and in drinking water. However, the true value of these data will only become apparent when we quantify the risk they pose to consumers. A drinking water risk assessment for organic micropollutats using the results of several independent studies has not yet been carried out in our country.

RESEARCH HYPOTHESES

After reviewing the scientific literature on the topic and defining my research objectives, I formulated the following hypotheses:

1. It can be demonstrated that the stable isotope ratio of H/D and $^{16}O/^{18}O$ can be used to determine the filtered water ratio in the wells of the riverbank filtered aquifer of Baja, and thus this method can be used as a substitute for the modelling-based determination.

2. My assumption is that the concentration of organic micropollutants in Budapest's riverbank filtered aquifers does not currently reach a critical level that would pose a risk to the drinking water consuming population, so Budapest's drinking water is safe to drink in terms of organic micropollutants.

3. It can be assumed that the group of the population that is present on social media platforms and obtains at least part of its information on drinking water quality from online media, considers the extent of the risk of organic micropollutants to drinking water supply and human health to be significant and fears the adverse health effects of organic micropollutants in drinking water.

RESEARCH OBJECTIVES

My research is mainly related to a better understanding of the processes of riverbank filtered drinking water aquifers. My research has mainly been conducted on the Baja aquifer, which has been in continuous operation for forty years. Although the aquifer has been in operation for quite a long time, it has not been the site of any scientific research, so I was the first to carry out some of the studies. In my research I also used data from Budapest aquifers published in the literature.

As the first step of my work, I defined my research objectives:

1. Determination of the ratio of H/D and $^{16}O/^{18}O$ stable isotopes in the riverbank filtered water of the Baja aquifer. Such any study has not been carried out in this aquifer so far, only model calculations based on hydrogeological studies have been used as a base for the classification.
2. Modelling of bank filtration processes under laboratory conditions in a pilot model plant. The aim was to investigate the concentration of organic micropollutants.
3. Investigation of the presence and concentration of certain organic micropollutants in the bank filtered wells of the Baja aquifer and in the Danube. My aim was to observe the

changes of the concentration of the tested compounds during the process of bank filtration.

4. Preparation of a human health risk assessment of organic micropollutants for drinking water consumption based on water quality data from bank filtrated aquifers. If the objectives set out in 2 and 3 do not result usable data, I will continue the work using published data series.
5. To survey and evaluate the knowledge and opinions of the population on organic micropollutants. I compare the results of my risk assessment with the results of the survey.

My research was primarily based on studies that had not yet been carried out in the water aquifer under study.

RESEARCH METHODS

I conducted my research according to the predefined objectives in order to confirm or refute the hypotheses.

- In my research, I used a variety of research methods, both inductive and deductive, but with a strong emphasis on synthesising independent information.
- I studied and analysed the relevant national and international literature in detail, and examined the legislation and standards. For this purpose I mainly used databases and journals available online.
- I carried out tests in the wells of the aquifers included in the study to determine the ratio of bank filtrated water and the concentration of organic micropollutants.
- I have set up and operated a pilot laboratory equipment for modelling the processes of bank filtration.
- I made a risk assessment to define the risk of organic micropollutants to drinking water supply and prioritised the compounds under investigation based on the results.
- I conducted a questionnaire survey to assess the knowledge and opinions of the population on organic micropollutants.

BRIEF DESCRIPTION OF THE STUDY

1. Determination of the bank filtered water ratio in the wells of the Baja drinking water aquifer using the stable isotope method

The ratio of bank filtered water in the production wells can be accurately determined by examining the stable isotope ratios of $^{1}H/^{2}H$ and $^{18}O/^{16}O$. One of the field of my research was

the water aquifer of the city of Baja, therefore the determination of the bank filtered water ratio was also carried out there. For the study, I took samples from two production wells of different capacities, the Danube as a surface water body and a groundwater well further away from the river. The stable isotope ratios in the water samples were determined by mass spectrometry, and the results were expressed as $\delta^{18}O$ and $\delta^{2}H$ in ‰ relative to the internationally accepted standard for hydrogen and oxygen isotope ratios in water. From my results, it was found that the proportion of filtered water of surface origin in the wells is 64% and 81%, thus both of them are considered as coastal filtered wells under the medium water level of the Danube.

2. Modelling and coupling of bank filtration with reverse osmosis under laboratory conditions

During my research, I considered it important to set up and test a pilot model plant, which is most similar to the processes of an operating bank filtration aquifer. The experiment involved the long-term operation of a slow sand filter fed with surface water and a reverse osmosis filtration system. The objective was to investigate the effectiveness of the slow sand filter in reducing the concentration of microbiological organisms and some organic micropollutants. The system was operated for more than a year, during which time we investigated the variation of water flow and pressure values at different points in the system, the variation of microbiological parameters, and the occurrence and concentration of some organic micropollutants during filtration. Organic micropollutants were analysed on two times during the study period by an external laboratory. The presence of the investigated organic micropollutants could not be detected by the laboratory within the measuring range of the available analytical instruments.

3. Investigation of the occurrence of organic micropollutants in the bank filtered water aquifer of Baja

Similar to the stable isotope analysis, the organic micropollutants were also analysed in the production wells of the Baja aquifer. 5 sampling points were selected for the study. During my research, I did not have access to properly accurate analytical instruments for the precise detection and measurement of organic micropollutants, so I cooperate with an accredited analytical laboratory for the analysis of the samples I took. I was actively involved in the preparation and analysis of the samples. The water samples were analysed using GC-MS and HPLC-MS instruments. The presence of the selected organic micropollutants could not be detected within the measuring range of the available analytical instruments.

4. Risk assessment of organic micropollutants in Budapest drinking water supply

The risk assessment was carried out by determining the risk quotient (RQ) and developing the method. I used published data for the study, summarising the data on organic micropollutants in the bank filtered production wells of drinking water aquifers of Budapest. From the data, the highest measured concentrations were selected and calculated using the worst-case scenario principle. Interpreting the results, I found that in all cases the risk factor was several orders of magnitude below the risk value. The five organic micropollutants with the highest risk were cefepime, carbamazepine, diclofenac, and risperidone pharmaceuticals, and the pesticide methachlor ESA. To make the results easier to compare, I created a grouping system to classify the different organic micropollutants with risk factors RQ<1 into "low", "very low" and "negligible".

5. Questionnaire survey to assess the knowledge and opinions of the Hungarian population on organic micropollutants

The aim of my questionnaire survey was to assess the knowledge and opinions of the population about drinking water and organic micropollutants. The questionnaire was prepared online and distributed through social media platforms. The sampling was random, i.e. the questionnaire could be filled in by anyone. The survey was conducted over 7 days, during which time a total of 320 valid responses were received. The responses reflect a rather negative perception of the quality of domestic drinking water. It is also very clear from the results that the presence of organic micropollutants in drinking water and their impact on human health is also a concern for the respondents. A further result of the questionnaire survey is that the majority of respondents fear that water quality problems caused by organic micropollutants will worsen in the future.

SUMMARY OF THE RESEARCH

The main aim of my research was to investigate the relationship between changes in organic micropollutant concentrations and processes during riverbank filtration. The first step was to determine the origin of the water produced by the wells in the Baja bank filtration aquifer.

Based on literature data, the most appropriate approach was to investigate the ratio of stable isotopes. Such an analysis could not be carried out previously for this aquifer. In view of the results, the work was continued by determining the presence and concentration of certain organic micropollutants. The study was carried out on the one hand on a pilot equipment set up under laboratory conditions and on the other hand on the already mentioned operating water

aquifer. The pilot slow sand filter and the combined RO module, which simulates the processes of bank filtration, have been in operation for more than a year. During this period, a number of parameters, including changes in the concentrations of some organic micropollutants, were investigated. Similarly, I have investigated the presence of organic micropollutants in water samples collected from the production wells of the aforementioned operating aquifer. As the analytical methods and facilities available to me were not able to detect the presence of the pollutants, the next stage of my work, risk assessment, was carried out using literature data. I compiled previously published data to produce a risk assesment that quantifies the risk from organic micropollutants to the drinking water consuming population in Budapest. Given that the results showed that the risk from the tested organic micropollutants was below the critical RQ=1, I created categories to distinguish and compare low risk factors. Based on the results, I ranked the 5 organic micropollutants with the highest risk.

In the last phase of my research, I conducted a questionnaire survey among the population using social media platforms and online media consumption. Finally, I compared the results of the risk assessment with the experience of my questionnaire survey, which was conducted to assess the knowledge and opinions of the domestic population about organic micropollutants.

I have achieved the objectives of my research, and I have been able to prove all three hypotheses I set out at the beginning of my work.

SUMMARISED CONCLUSIONS

One of the most important findings of 20th century man is that the Earth's water resources are finite and anthropogenic impacts can significantly reduce their quality and lead to permanent degradation. Reducing consumption is therefore a priority, and great emphasis must be placed on protecting the quantity and quality of existing water resources. A thorough study of the groups of organic micropollutants shows that industry is producing more and more of these compounds every year, in response to the demands of consumer society. Research in recent years has shown that organic micropollutants of anthropogenic origin can be found in many parts of the world, although their measurable concentrations can vary widely. The problem is that, partly because of their diversity and partly because of the slowness of the legislative system, a significant proportion of these pollutants are still not regulated. With a few exceptions, most organic micropollutants are not part of routine monitoring programmes and there are no environmental quality or drinking water safety limits for them. Consequently, it can be concluded that a significant proportion of these pollutants are released to the environment without adequate controls. Without comprehensive monitoring programmes, we

cannot have sufficient information on the environmental fate of organic micropollutants. During my research, I felt it was important to have an accurate understanding of the origin of the water in the bank filtered aquifer I was studying. Therefore, I determined the amount of stable isotopes ^{2}H and ^{1}H, as well as ^{18}O and ^{16}O for two wells of the Baja aquifer included in my research, and calculated the proportion of bank filtered water using the data obtained. From my results, it was found that the wells have 64% and 81% filtered water from surface origin, respectively, so both can be classified as riverbank filtered wells. The stable isotope analysis method provided useful information on the water ratio in the bank filtered wells, allowing me to refine the findings of the modelling calculations.

I investigated the presence of organic micropollutants in the raw and filtered water of a laboratory model plant and in the wells of the operating aquifer. In the water samples I took, I could not detect any organic micropollutants in the range tested. This led me to conclude that the selected compounds may be present at concentrations below the detection limit of the analytical method available to me (0.05-0.5 ng/ml).

I was able to support my conclusion with published water chemistry data from the Danube and the Budapest section of the river. Indeed, some of the organic micropollutants that I also studied were present in some of the bank filtered wells, typically in the lower range of 0.005-0.29 µg/l. In the river section below Budapest, I assumed a decrease in the concentration of these compounds, since the number of discharges and the volume of effluent discharged is lower than in the Danube section of the capital. In natural waters, degradation of organic micropollutants is typical.

In my thesis, I have presented in detail the method of risk assessment of organic micropollutants that I have applied in the drinking water supply and in the calculation of the impact on the health of consumers. The formula takes into account factors such as body weight, tolerable daily intake, average daily tap water consumption, gastrointestinal absorption and frequency of exposure. For the calculation of the risk factor, the worst case scenario principle was applied, i.e. the maximum concentration values for each organic micropollutant were used. Interpreting the results, I concluded that the risk factor (RQ) was in all cases several orders of magnitude below the critical value of 1. The risk factor was highest for cefepime, metazachlor ESA, carbamazepine, diclofenac and risperidone. To make the results easier to compare, I have created a grouping system to classify the different organic micropollutants with risk factors RQ<1 into "low", "very low" and "negligible". I compared my results with data on the effectiveness of riverbank filtration. It can be declared that the risk of the antidepressant

carbamazepine is high, with a very small decrease (4.2-6.4%) in its concentration during the process of riverbank filtration.

Despite my results above, I assumed that the domestic population is pessimistic about the risk of organic micropollutants to human health, i.e. that the public fears organic micropollutants to a much greater extent than can be supported by risk assessment. To verify my hypothesis, I conducted a questionnaire survey to assess the opinions and knowledge of the domestic population about organic micropollutants. The survey focused on the population using social media and obtaining information largely from online media. From the responses, I concluded that the presence of organic micropollutants in drinking water and their impact on human health is a cause for concern among those who completed the questionnaire and that many people fear the adverse health effects of organic micropollutants in drinking water.

NEW SCIENTIFIC RESULTS, THESISES

1. I was the first to carry out a measurement of the stable isotope ratios of $^{1}H/^{2}H$ and $^{16}O/^{18}O$ in the Baja bank filtered drinking water aquifer, and based on this I determined the ratio of the wells' surface orgin filtrated water. This proved that the method can be used to replace and refine the previous model-based determination in the case of the aquifer under investigation. The phenomenon of clogging that may occur during long-term operation of the bank filtered aquifer can be determined and predicted by regular analysis of the stable isotope ratio of the produced water.
2. My results confirm that in the section of the Danube near Baja, the organic micropollutants I have analysed are typically present at low concentrations, below the detection limit of the available laboratory analytical tools. The risk assessment was carried out by summarising and processing published data and data series for the bank filtered drinking water sources supplying Budapest. The risk factors calculated for each organic micropollutant were typically several orders of magnitude lower than the critical values calculated based on literature data. By developing this method, a classification system was developed to group risk factors RQ<1 and to categorise each organic micropollutant as 'low', 'very low' or 'negligible'.
3. By analysing the results of my questionnaire survey of the domestic population using social media, I found that the group of the domestic population I surveyed perceives the extent of the risk of organic micropollutants to drinking water supply and human health as significant and fears the adverse health effects of organic micropollutants entering the body from

drinking water. I have also been able to confirm that respondents expect the level of risk from these pollutants to increase, i.e. that the population fears a further deterioration of the current situation.

PRACTICAL USE OF RESEARCH RESULTS

The stable isotope geochemical analysis presented in my research is not part of the routine tasks performed in the operation of bank filtered aquifers. The knowledge of the filtration water ratio is of great importance in the case of bank filtered wells, so it would be worthwhile to include this test in the practical tasks and to perform it several times under the varying hydrological conditions of the surface water body of the water body. This would add valuable data to our knowledge of riverbank filtered aquifers.

In my dissertation, I was looking for answers to the question of the risk of organic micropollutants to drinking water consumers. Such a comprehensive risk assessment and analysis has not been and could not have been carried out until now, because detailed and credible data on organic micropollutants have only been produced in recent years. I collected and aggregated these results and used the resulting data set to identify risk factors. I was thus able to give a view of the current extent of the problem of organic micropollutants. I found that although organic micropollutants may pose a risk to human health at higher concentrations, at their current measurable concentrations this impact is very low, several orders of magnitude below the critical level. The results of my risk assessment could be used in a more comprehensive, possibly country-wide, study of the presence and risk of these pollutants. On the other hand, my findings may help future similar studies as a reference.

I drew attention to the fact that the knowledge of the Hungarian population about the drinking water supply and the extent of the risk from organic micropollutants is insufficient. There is a need for information and education, either in print or online, in the form of multimedia, to bring the population's knowledge of this issue closer to reality. There is a need for a rapid and clear response from the professionals to the often inaccurate articles on the subject.

RECOMMENDATIONS

I recommend the results of my thesis primarily to professional organisations such as the Hungarian Water Utility Association (MaVíz) or the Water Management Directorates. The results of the research and studies carried out in my thesis may be helpful for the studies and future work of universities, research institutes and research groups. The risk assessment method I have presented is simple and easy to apply, and could therefore be used in a larger, more

comprehensive research project. I recommend the use of the risk assessment method presented in my thesis to all researchers and laboratory staff involved in the study of organic micropollutants. The comments I have made can serve as a base for reviewing and revising current legislation, regulations and standards. I recommend my thesis as a supplementary teaching material for teachers and students of higher education institutions working in the field of water science. My thesis can be useful for professionals in the field of environmental protection and drinking water safety who, like me, consider it a priority to preserve the status of our water resources and drinking water sources for posterity.

LIST OF PUBLICATIONS OF THE CANDIDATE

Articles published in a peer-reviewed journal:

Salamon Endre, Goda Zoltán, Berek Tamás: *Analysis of reverse osmosis filter permeability.* Pollack Periodica An International Journal for Engineering and Information Sciences, 2018. Vol. 13, No. 3, pp. 221–230 ISSN 1788-3911

Salamon Endre; Goda Zoltán: *Coupling Riverbank Filtration with Reverse Osmosis May Favor Short Distances between Wells and Riverbanks at RBF Sites on the River Danube in Hungary.* Water, 2019, 11: 1 pp. 113-124.

Goda Zoltán: *Hazai üzemelő és távlati parti szűrésű ivóvízbázisok mennyiségi és minőségi értékelése.* Hadmérnök, 2019, 14:2 pp. 157-166.

Goda Zoltán: *Szerves mikroszennyezők kockázatelemzése a parti szűrésen alapuló ivóvízellátásban.* Hadmérnök, 2021 16(1), 79–94.

Goda Zoltán: *Az éghajlatváltozás hatásai az oldott oxigén koncentrációjára parti szűrésű vízbázisokban.* Hidrológiai Közlöny, 2021. 101(4): p. 18-25.

Books, book chapters:

Goda Zoltán: *Szerves mikroszennyezők kockázatelemzése a vízi környezetben és az ivóvízellátásban.* In: Hausner Gábor (szerk.): Szemelvények a katonai műszaki tudományok eredményeiből II., Ludovika Egyetemi Kiadó, 2020. Budapest. pp. 118-134., ISBN 978-963-531-441-6

Goda Zoltán: *Szerves mikroszennyezők előfordulása ivóvízbázisokban.* In: Knisz, Judit (szerk.): Szerves mikroszennyezők a vizekben. Budapest, Ludovika Egyetemi Kiadó (2020) pp. 107-132., ISBN: 9789635313624

Goda Zoltán: *Szerves mikroszennyezők kockázatbecslése.* In: Knisz, Judit (szerk.): Szerves mikroszennyezők a vizekben. Budapest, Ludovika Egyetemi Kiadó (2020) pp. 133-155., ISBN: 9789635313624

Goda Zoltán: *A vízszerzés módjai és műtárgyai.* In: Vadkerti Edit (szerk.): Vízszerzés, víztisztítás. Budapest, Ludovika Egyetemi Kiadó (2021) pp. 11-42., ISBN: 9789635314492

PROFESSIONAL-SCIENTIFIC CURRICULUM VITAE OF THE CANDIDATE

Name: Zoltán Goda

Place and date of birth: 12. November 1979, Baja, Hungary

Studies:

In 2002, he graduated from the Faculty of Technology of the Eötvös József College in Baja, Hungary, with a degree in Environmental Engineering, and in 2007 from the Faculty of Chemical Engineering of the Budapest University of Technology and Economics, Hungary.

Professional career:

From 2003, he worked at Baja and Térsége Víz- és Csatornamű Kft. as a water utility network registrar, but his duties also included the company's environmental and waste management tasks and responsibilities. From spring 2007 until 2016, he continued his work as the plant manager of the Baja waterworks. In addition to the day-to-day management of the waterworks, he was also responsible for monitoring the operation of the riverbank filtered water aquifer and coordinating the aquifer protection tasks. In 2015 and 2016, he managed the trial operation of the new water treatment technology during the drinking water quality improvement project, which involved the complete reconstruction of the waterworks. In order to ensure the quality of his duties, he obtained a fire safety qualification for the use of explosive materials.

From 2016, he worked at the Eötvös József College as a technical coordinator, and after the transformation of the institution, the Faculty of Water Sciences of the National University of Public Service also counted on his work. Her main tasks included the technical coordination of several international research projects at the institutional level, but he also participated in the implementation of national proposals and programmes involved by the University. In connection with these tasks, he also undertook several study tours abroad. He is currently working as a research assistant at the Faculty of Water Sciences. In addition to his research

activities, he is also involved in teaching, specialising mainly in drinking water abstraction and treatment and meteorology. His teaching activities also include foreign language teaching.

Language skills:

He has a complex level (C1) in English and a basic level (B1) in Italian.

Scientific activity:

During his studies, the candidate successfully applied for a 12-month grant from the New National Excellence Programme (ÚNKP) to study the origin and oxygen balance of the water in the Baja bank filtered aquifer. He is actively involved in the Hungarian scientific life, being a member of the Water Supply Section of the Hungarian Hydrological Society and the Climate Section of the Hungarian Meteorological Society.

Budapest, 16 February 2023.

Zoltán Goda