Complex health risk assessment of drinking and bathing water

Thesis

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Introduction

The natural and built environment (both outdoors and indoors) has major impact on human health. The effects may be physical, chemical or biological by nature, and positive or negative. The importance of healthy environment is recognized by national and international legal agreements. In Hungary, the Fundamental Law states the right to healthy environment for everyone. Of the 17 Sustainable Development goals, which were defined by the United Nations for the period 2015-2030, three are directly, and almost all of them are indirectly related to healthy environment. The health impact of environmental factors, and the mitigation or reduction of adverse effects is the main focus of the European Health and Environment Process launched by the World Health Organization in 1989, and the Interministerial Conferences on Environment and Health which are organized in its framework.

A significant proportion of environmental health risks relates to water for human use, especially drinking and bathing water. The General Assembly of the United Nations recognised the access to drinking water as a basic human right in 2010, implying that everyone in the world has right to sufficient, safe, acceptable, accessible and affordable water. Universal access is presented as a stand-alone goal among the Sustainable Development Goals.

For centuries, both drinking water and bathing water was associated with significant microbiological risk which comprised a major part of the burden of disease. The means of risk management were mostly the application of different water disinfection materials and technologies. As a result, the frequency of waterborne outbreaks was reduced to a very low level, starting from the early-mid 20th century. Nowadays, extensive enteric outbreaks are mostly associated with disaster situations. However, the prevalence of sporadic water related infections cannot be ignored. The methodology of quantitative risk assessment is useful to quantify both the risk and the efficiency of risk management interventions.

The prevention of water borne diseases, or the fear from such infections often prompts the overuse of water disinfectant chemicals and technologies (e.g. overchlorination) which, as we know now, can in turn lead to a significant increase in chemical, especially carcinogenic risks. Besides disinfection, the other method to mitigate the risk from consuming contaminated shallow groundwater was the drilling of artesian well from the 1930s. These wells, however, often contained chemical contaminants in concentrations above the health safety limit, though this fact was only discovered in the 1980s. In the past decades, the geogenic chemical contaminants presented the highest risk in Hungary, affecting almost 1.5 million people. A national Drinking Water Quality Improvement Programme was launched after the accession to the European Union to solve this problem, with extensive resource allocation. Though the programme was successful in improving this aspect of drinking water quality, the focus of investments was compliance with the legal parametric value, the health risk assessment based, complex approach was missing. Iodine is also a geogenic compound, but it deserves attention more as a deficient nutrient, than a contaminant.

Thought health effect of disinfection by-products and its extent is comparable to geogenic contaminants, comprehensive programmes or plans to control the former are missing, and the legal regulation is incomplete, and not health-focused. As a consequence, the methodology for the detection of contaminants is also insufficiently developed. The high risk to vulnerable groups, especially infants and toddlers, which can exceed that of the general population by another of magnitude, increases the significance of the problem. Though drinking water is usually considered higher risk because of direct consumption, in the case of disinfection by-products bathing water has similar importance, due to its exposure characteristics.

Emerging contaminants are a new challenge. Some of them are similar in composition and mechanism of action to previous compounds, but some of the new chemical groups, such as endocrine disruptors or nanomaterials require a completely new approach in health risk assessment.

The risk-based approach is used increasingly in water safety. It requires a new, holistic perspective, covering different types of risks from different sources. To evaluate concomitant risk (such as microbiological risks and disinfection by-products) and to identify priorities, quantitative risk assessment is indispensable.

Objectives

Complex health risk assessment of water for human use, with special focus on parameters having high health relevance in the Hungarian population. In this framework, the health impact assessment and risk assessment of chlorination by-products, and its comparison to microbiologial risks and geogenic pollutants (Figure 1):

- Microbiological risk assessment:
    - Evaluation of microbiological non-compliance in Hungarian drinking and bathing waters
    - Assessment of a drinking water outbreak related to an extreme weather event
    - Evaluation of the vulnerability of drinking water supplies and the risk assessment of intentional water pollution (case studies)
    - Health risk evaluation of pools operating without disinfection
- Risk assessment of chlorination by-products
    - Drinking water
        - Health risk assessment of chlorination by-products
        - Identification of sensitive sub-populations in accordance with the current international and national guidelines
    - Bathing water
        - Health risk assessment of chlorination by-products
        - Identification of the main exposure matrix and risk groups
        - Comparison of the water-air concentration speciation models of the risk assessment softwares and measurement data
- Health risk assessment for the major geogenic pollutants in Hungary
    - Health risk assessment of arsenic
    - Health risk assessment of iodine
- Comparison of health impact from microbiological, anthropogenic chemical (chlorine disinfection) and non-anthropogenic, geogenic pollutants and their complex evaluation.
- Evaluation of risk assessment possibilities for emerging contaminants (nanomaterials).

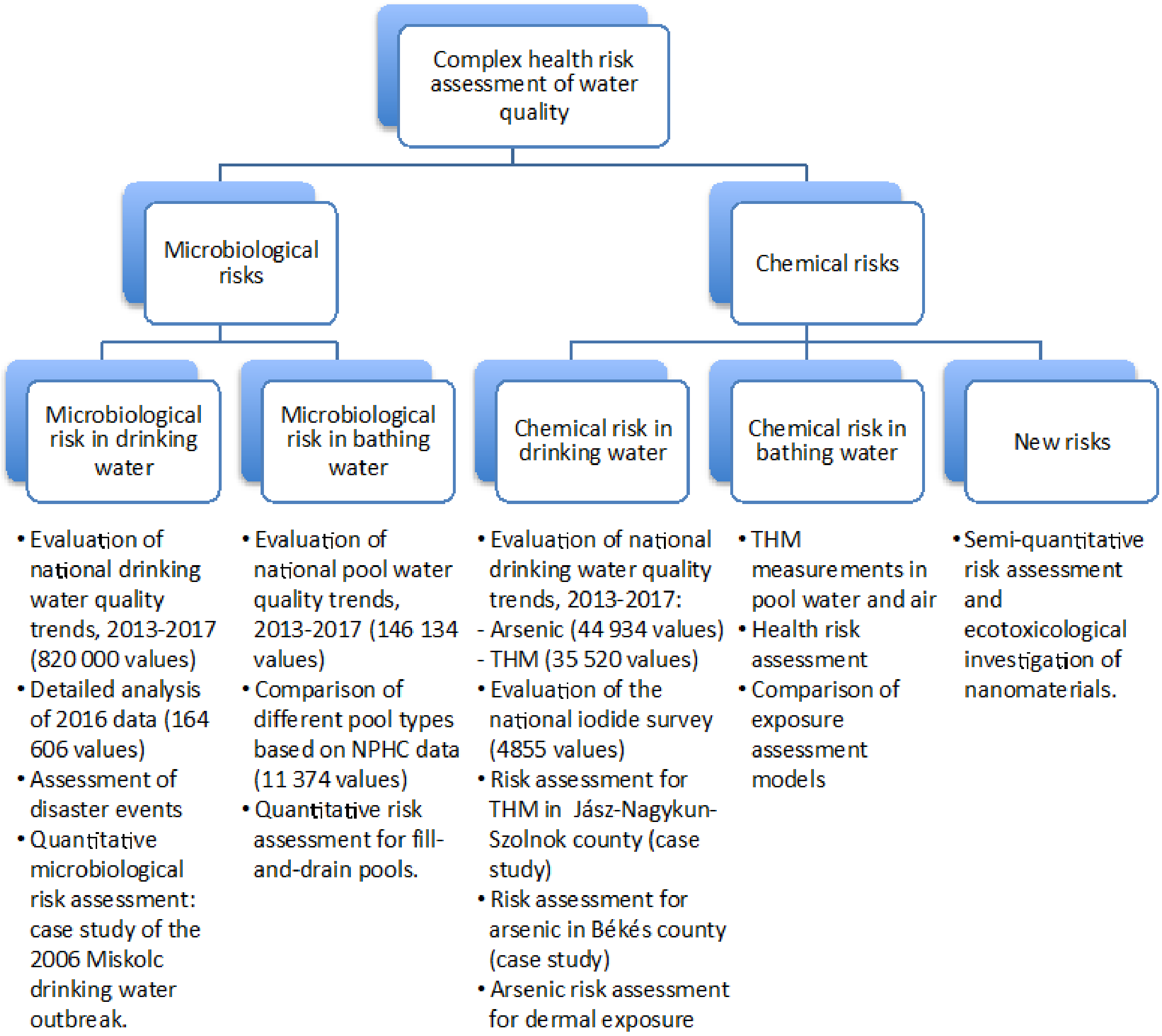

Figure 1: Overview of measurements and evaluations included in the PhD study for complex health risk assessment.

Materials and Methods

Drinking water quality monitoring

The evaluation of national drinking water quality was based on the data from routine monitoring. The quality of drinking water is monitored by the water suppliers and the competent public health authorities according to the Government Decree 201/2001 (X. 25.) on „the quality and monitoring requirements of drinking water". Results are uploaded to a central online database managed by the National Public Health Centre (NPCH), the Information System of Water in Human Use (HUMVI). Data from the Information System (50.000 samples and 500.000 measurements per year) aggregated for the municipalities in the period 2013-2017 was used to assess drinking water quality on the national level, considering the following parameters: *E. coli, Enterococcus,* coliforms, colony count 22 °C, arsenic, chlorination by-products. More detailed spatial analysis was carried for data from 2016. Microbiological parameters and trihalomethanes (THMs) were characterized by the percentage of compliant samples, arsenic concentration by the yearly median value. Compliance was evaluated against the legal parametric value (Government Decree 201/2001 Annex 1).

Microbiological quality of pools and spas

Pool water quality is monitored by the pool operators and the district public health authorities in accordance with the Ministerial Decree 37/1996 (X. 18.) of the Minister of Welfare on the establishment and operation requirements of public baths. The national data collection does not applies to pools, thus the source of data were the yearly report from the public health offices for 2013-2017 (8000 samples a year), and the results of pool water analysis performed in NPCH as an analytical laboratories (1800 samples). The former was used for the representative national assessment, while the latter for the determination of bacterial count ranges which pose a risk to health.

National pool water quality data is presented in county level aggregation. Limit values from the classification standards were used for the evaluation of analytical results. Differences in pool water quality were analysed by the mode of operation (fill-and-drain or water circulation) and pool type.

Chlorination by-products in pools

A targeted series of measurements was carried out to investigate indoor air quality in pools. Altogether, 19 swimming pools and 3 whirlpools were analysed in 2015-2016. At the study sites, the parameters of water treatment and pool operation, technical characteristics, means and frequency of ventilation were recorded. Water samples were collected from feed water and pool water. Water temperature, pH and conductivity were measured on site. The concentration of absorbable halogenated compounds, free and combined active chlorine, and THMs was also determined. Active sampler was used for the collection of air samples at 40 and 150 cm above the water surface, corresponding to the inhalation height of swimmers and staff in the pool area.

National iodine survey

National survey was carried out in 2016 to determine the iodine concentration of drinking waters. Drinking water samples were collected in every drinking water supply zone, and iodine content was determined by ion chromatography. The results were extrapolated to all municipalities.

Risk of nanomaterials

Iron(II,III) oxide nanoparticles (average diameter 29 nm) were tested by ecotoxicological methods for risk assessment. *Daphnia magna* acute and chronic tests, *Thamnocephalus platyurus* acute tests, and growth inhibition tests were applied, using *Navicula pelliculosa* (diatome) *Anabaena* sp. (cyanobacterium) and *Pseudokirchneriella subcapitata* (green alga) test organisms.

Statistical analysis

Correlation was tested using Pearson linear correlation coefficient (r) analysis), with paired significance test (p). Independent groups were compared by Mann-Whitney test. IBM SPSS Statistics for Windows, version 21 (IBM Corp., Armonk, NY, USA) software package was used for the analysis.

Chemical risk assessment

Health risk of chemical contaminants in water for human use was determined using quantitative risk assessment. Direct toxic (acute and chronic) and carcinogenic risk was assessed separately. In the current study, carcinogenic risk of arsenic via drinking, carcinogenic and non-carcinogenic risk of bathing/showering, carcinogenic risk of THMs and non-carcinogenic risk of iodine were analysed for drinking water. The carcinogenic risk of THMs in pool water and pool air was assessed for swimming and indoor presence in pools.

Drinking water risk assessment was carried out using ”Risk-Integrated Software for Cleanups version 4” (RISC4) software. Human exposure parameters were obtained from the software and the US EPA Exposure Handbook. Variability of risk values was reduced using Monte Carlo simulation. Two models were compared for the analysis of risk associated with pools. Chloroform was used as a model compound for the calculations. SWIMODEL is designed specifically for the the assessment of risks associated with swimming. Besides oral, inhalation and dermal exposure it also takes into account the uptake through mucosa (buccal, sublingual, nasal and aural). The other applied model was the web-based ConsExpo. The „Disinfectant Fact Sheet was used in accordance with the guidance of the developer Dutch Research Institute (RIVM).

Quantitative microbiological risk assessment

The Miskolc drinking water outbreak of 2006 was used as a case study for the evaluation of the quantitative risk assessment methodology. The research questions were the following: whether the norovirus titre detected in drinking water (25-250 GC/l) explained the observed number of infection; what level of protection do the different risk management interventions (boiling, increased chlorination, ultrafiltration technology). The basis of calculation was the lower consumption of untreated water (250 ml) for boiling, lower infective dose in case of elevated chlorine level (0,4 mg/l and 0,8 mg/l) and high efficiency virus removal ($>10^6$) for ultrafiltration. Dose-response curve was calculated using beta-Poisson model.

For pools, the yearly *Cryptosporidium* infection risk in fill-and-drain pools was calculated. *Cryptosporidium* is the etiological agent of most bathing water related infections. Though bacterial indicators do not indicate adequately the presence of *Cryptosporidium* in disinfected pools due to their different chlorine sensitivity, in non-disinfected pools a correlation between faecal indicator count and enteric pathogens is implied ($1: 10^6$). Though swimming is not general practice in fill-and-drain pools, the same ingested volume of water per bathing event (32 ml) was used for the calculation as for swimming pools, to represent the worst case scenario. Inactivation was not taken into account in the absence of disinfection, only the effect of dilution was calculated. The risk of illness after infection is 70 %. Exponential dose-response relationship was used for *Cryptosporidium.*

Results

- Microbial quality of drinking water was characterized by 1-2 % non-compliance for faecal indicators, and higher exceedance rate (4-6 %) for other indicator organisms. However, there is significant spatial and temporal variation behind the seemingly reassuring results. Non-compliance is observed with much higher frequency in small community water supplies, in relation to their presumably lower infrastructural and operational background. Altogether, the main obstacle to indicator-based health risk assessment is the insufficient data collection. Indicator-based classification does not provide an objective picture on microbiological quality of drinking water due to the nature of the monitoring practice (timed and planned sampling, in smaller water supply zones only four times a year).
- The case study of the 2006 Miskolc drinking water outbreak confirmed the usefulness of quantitative risk assessment in determining health risk for larger scale drinking water contamination and the evaluation of risk mitigation. Boil water advice reduced the risk by less than one, while chlorination by approximately 2 orders of magnitude, but the risk of infection was above the tolerable level even in the most favourable scenario. Ultrafiltration is a suitable solution to provide microbiologically safe water, even if a similar contamination event occurs than in 2006. Ultrafiltration reduced the risk of infection well below the acceptable level.
- The analysis of health risk in bathing water revealed a high level of microbiological compliance in circulated and disinfected pools, while among the fill-and-drain pools the proportion of fully compliant pools is only 30-40 % and 30 % is severely contaminated. These ratios are the same for decades, indicating a systematic problem of operation, regulation and control. Comparing the calculated risk to that of the circulated pools, the geometric mean and the maximum risk is more than one, the 95 % (which is the most relevant value for risk characterization) is more than two orders of magnitude higher. The difference is even better illustrated by the ratio of pools where the yearly risk of infection is higher than the tolerable level of 10^{-4}: it was almost 50 % (106/218) of the fill-and-drain, and 2.3 % (16/684) of the circulated pools.
- Inhalation is the most significant risk for chlorination by-products. For sensitive sub-populations, even THM concentrations below the regulatory limit value do not guarantee sufficient health protection. Both risk assessment models confirmed that inhalation in itself results in excess carcinogenic risk of the exposed population exceeding the tolerable level.

Dermal uptake is the second most important pathway after inhalation. The excess risk from cumulative life-time (child 7-10, child 11-14 and adult) dermal exposure is also close to the acceptable level. In pools, the THM concentration in air estimated by the exposure models differs from the measured values by several orders of magnitude. Thus regulation and monitoring should be based primarily on air quality measurements.

- Health risk assessment of geogenic pollutants indicated high excess carcinogenic risk in the selected study area for the initial period (1980-1990), more than 100-fold above the tolerable level. The potential risk is mainly derived from drinking water consumption, dermal uptake is negligible in comparison. The consumption of locally grown produce irrigated by arsenic contaminated tap water is also insignificant compared to drinking. The risk estimated from the characteristic concentration (24 μg/l) between 1991-2011 is still significant, due to the long, two decades of exposure. The daily intake calculated from the 2017 arsenic concentration in drinking water is 0.07 μg/bw kg, implying two orders of magnitude lower risk level.
- The national survey shown relatively stable concentration of iodide in the drinking water distribution system, thus the concentration within a drinking water supply zone is considered uniform. The iodide level was above the limit of detection only in a fraction of the investigated municipalities. Looking at the municipal level data, the concentration of iodide was detected, but below the recommended intake level in 64 municipalities. The iodide requirement of children is higher, thus drinking water contributes even less in meeting it. With the exception of 1-6 years old children, not even the highest concentration exceeded significantly the tolerable intake range.
- The biological impact of nanoparticle contaminants, as an emerging risk, is significantly different from the normal size particles. Ecotoxicological methods are suitable for the characterization of this difference while modelling is used to differentiate their impact.

Conclusions, thesis

- The microbiological quality of tap water is very good on a national level, but the aggregated compliance rates mask the significant differences between geographical areas and different size water supplies. Due to the low number of samples, endpoint monitoring is not sufficient for the prediction of infectious risk, as the Miskolc outbreak well illustrates. The solution is the introduction of preventive, risk-based methods, such as the early warning system relying on turbidity which was successfully introduced in Miskolc. The importance of these methods is particularly high for the vulnerable drinking water resources which are more exposed to effect of extreme weather events increasing with climate change.

- Microbiological load of non-disinfected fill-and-drain pools is multiple times higher than circulated pools. The yearly risk of enteric infection is more than 10^{-3}, well above the tolerable level. Thus the revision and update of pool and spa regulation is indispensable, finding suitable means to reduce infectious risk in case of therapeutic waters as well.
- Disinfection, while reducing microbiological risks, generates a new chemical hazard. The adverse health effect of disinfection by-products was unambiguously confirmed for both drinking and bathing water exposure. The extent of risk cannot be directly compared to the infectious risk, due to the differences in prevalence and health outcomes. The calculation of disability adjusted life-years would be a potential tool for direct comparison. The Hungarian THM regulation is strict in international comparison. Therefore the elevation of the limit values is a frequent request from water suppliers and pool operators. The presented results indicate that this is not feasible, particularly because of the sensitive sub-populations (children). Moreover, the current regulatory value does not provide sufficient health protection in pools, especially for elite swimmers. The formation of disinfection by-products should be reduced without compromising disinfection efficiency, e.g. by requirements on reducing organic matter content.
- Risk assessment of chlorination by-products is leads to different results depending on the model used for the estimation concentration in air. Indoor air quality of pools should be therefore monitored directly by air sampling, it cannot rely solely on the result of water quality analysis.
- The health risk of the most relevant geogenic contaminant, arsenic, was reduced significantly as a result of investments for its mitigation. However, water quality parameters cannot be handled separately: many municipalities faced new drinking water quality problems (e.g. *Pseudomonas* colonization or non-compliance of disinfection by-products) after the Drinking Water Quality Improvement Programme, which may be comparable to the risk from arsenic. This finding underlines the importance of integrated risk management, one of the key elements of risk based drinking water safety methodologies.

- Low iodide concentration in the majority of drinking waters in Hungary supports the need for iodine supplementation in most regions of the country. Only 4 municipalities can safely derogate from the general guidance on the use of iodinated salt.
- Further investigation is necessary for the health risk assessment of emerging risk factors, such as the nanomaterials. Their introduction in drinking water treatment should be deferred until sufficient evidence is available for their safe application.
- Hungary is leading the way in regulating the implementation of risk based approach, drinking water safety planning is an obligation since 2017 in all water supplies. However, the current methodology is only descriptive or semi-quantitative. Quantitative methods are not used even for those drinking water contaminants, where the necessary background data (e.g. environmental concentration, toxicological profile). The presented calculations indicate the value of chemical and microbiological risk assessment in the comparative evaluation of risks and the characterization of intervention efficacy.

Publications related to the thesis

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