

Risk analysis for groundwater intake in an old mining shaft with increased chloride content, Upper Silesia, Southern Poland

Beata Kończak and Ewa Janson

ABSTRACT

Water from underground intakes is the main source of potable water for people in Poland, hence the protection of its resources is of great importance for the functioning of society and the economy. A new regulation in Polish Water Law imposes the obligation of performing risk analysis for water intakes, including the assessment of health hazards, factors negatively affecting water quality which are identified on the basis of hydrogeological and geological analyses. The main objective of the study was to determine the health risk for chlorides and to present an innovative approach to the health risk for non-toxic substances. In Upper Silesia, which is the most industrialized and urbanized area in Poland, old mining shafts are often used as deep wells in the water supply chain, and higher mineralization is the key feature of abstracted water which does not quite eliminate them as a source of drinking water supply. This paper proposes a new method of health risk determination as hazard index (HI). We present analysis of the health risks with increased concentration of chlorides in water which cause health effects for water consumers, especially for men, children aged 4–8, pregnant women and women during lactation.

Key words | drinking water quality, groundwater intakes, health hazard, old mine shaft, risk analysis, water resources protection

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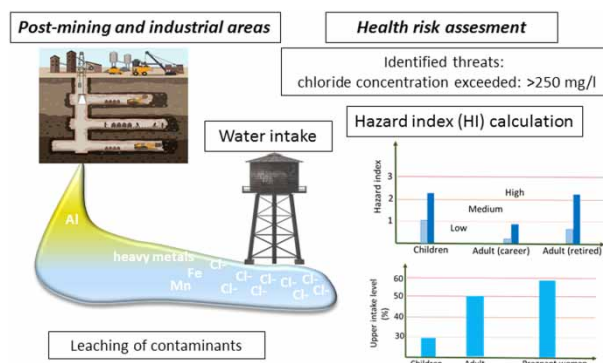
HIGHLIGHTS

- Children and the elderly are the most threatened by consuming chloride-rich water.
- Mining water causes an increase in the chloride concentration of drinking water.
- The extended health risk assessment should be done for the water intake localized on mining or industrial area.

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GRAPHICAL ABSTRACT



INTRODUCTION

The Upper Silesian Coal Basin, Southern Poland, is a region which is affected by mining activity which has been carried out for 200 years and is now in a transition process. Mine closure, restructuring of the infrastructure and the beneficial use of abandoned mines are part of a wider mandate for the mining industry to move towards long-term environmental and socio-economic sustainability (Petritz *et al.* 2009).

Old mine shafts are often used as wells for drinking water supply, especially those in relatively water-rich areas where permeable Quaternary strata cover Carboniferous aquifers. The owners of these intakes who perform tasks in the field of water supply must comply with the quality standards of water with regard to increasing the wellbeing of society and its demand for high quality services. In addition, legal requirements, technological progress and the principles of sustainable development force the service providers of the water supply to take many actions to ensure the primary objective, i.e., continuous water supply of very good quality for human consumption.

Thus, use of water from old shafts for drinking purposes is one benefit of the transition of a coal region, but on the other hand, problems of quality and an increase in some parameters, i.e., chloride content, limits the use of water from shafts without the use of technological solutions or risk assessment for the stability of the water chemistry.

REGULATORY CONTROLS FOR DRINKING WATER FROM GROUNDWATER INTAKES

Technological and legal solutions are conducive to ensure the supply of the population with drinking water of the required quality and in the desired quantity. The entrepreneurs providing water have an impact on many factors, such as controlling the processes of water treatment, distribution, supply and monitoring of these processes. There are also legal and economic instruments for protecting water intakes. Recently, in Poland, there have been major changes in the legislative field, which brought into force the provisions of the Water Law Act of 20 July 2018, which are currently in line with the EU Water Framework Directive (2000/60/EC). One of these requirements, which concerns the protection of water intakes, is the obligation to carry out risk analysis for water intakes. This analysis, in accordance with the requirements resulting from the new regulations, should assess health hazards, including factors which negatively affect the quality of the water from the intake. Identification of these factors is to be carried out based on hydrogeological or hydrological analyses (depending on the type of intake), water quality analysis, as well as the identification of hazards resulting from the land use as well as urban and industrial impact. It is worth mentioning that this obligation is new in Polish water management. The results of the risk analysis should lead to the justification of

the establishment of a protection zone, which will have specific orders, prohibitions and restrictions in the scope of land use and water use. Carrying out such analysis and then establishing a protection zone for water intakes will require the establishment of local law acts along with the implementation of restrictions, prohibitions and orders in the area of the protection zone. The impact of human activity on the natural environment, and the quality of groundwater and surface water is visible, especially in highly industrial and urban areas. In Polish legislation, only risk analysis for water intakes are obligatory, while such analysis is part of a wider water safety management tool: Water Safety Plan (WSP). WSPs have an important role to improve water safety for small systems, which commonly face challenges related to human and financial resources, training, equipment, geographic remoteness and highly variable water supply system types and management arrangements (WHO 2017).

In the European Union (EU), Directive 2015/1787 of 6 October 2015 amends Annexes II and III of the EU Drinking Water Directive, giving EU member states (among others) the option to deviate from the list of drinking-water monitoring parameters and from the stipulated minimum monitoring frequency in case a risk assessment has been implemented as a basis for the deviation.

The Protocol on Water and Health to the 1992 Convention on the Protection and Use of Transboundary Watercourses and International Lakes provides a legally binding framework for the WHO European region that requires countries that have become a party to establish national targets to achieve or maintain a high level of protection from water-related diseases. In this context, several countries have set targets for safe management. For example, Norway undertook to have a satisfactory internal control system by 2016 that includes a risk and vulnerability analysis that considers the effects of climate change for all water and sewerage works that serve 50 persons or more. Serbia undertook to develop legislation for the implementation of WSPs, and the Republic of Moldova endeavoured to have WSPs for all cities by 2015, and for all other settlements serving more than 5,000 people by 2020. In comparison, in Poland, the provisions of the new Water Law require the analysis of the health risk for water intakes with an amount of more than 10 m³ per day or supplying

more than 50 people (therefore, practically all water intakes), if water is supplied for human consumption as part of commercial, industrial or public buildings (Water Law Act of 20 July 2017, Journal of Laws of 2017, item 1566, Article 133, Article 138).

In Poland, WSPs are not yet required. Where WSPs are legally required, external audits (generally formal) are necessary to confirm compliance with relevant WSP requirements. In England and Wales, for instance, national drinking-water regulations require water suppliers to implement WSPs, and the Drinking Water Inspectorate (DWI) is charged with WSP auditing as part of the enforcement of the regulations. In addition, to confirm regulatory compliance, the audit process helps water suppliers to strengthen their WSPs by addressing improvement opportunities identified by DWI auditors.

Where WSPs are not legally required and where formal WSP audits are not appropriate, informal external audits have an important role to play. In Nepal, for example, the Department of Water Supply and Sanitation (DWSS) undertakes informal WSP audits for community-managed water supply systems (among other system types) in order to verify understanding of WSP principles, to discuss any barriers to WSP implementation. In Kenya, Uganda and the United Republic of Tanzania, informal WSP audits were carried out at three water utilities (one per country). Results from the audits revealed that 77% of the WSP process was well developed and implemented but gaps remained in operational monitoring and verification that could undermine WSP effectiveness (WHO 2017).

The audits further provided a mechanism to confirm required upgrades for ageing infrastructure to support the preparation of informed investment plans. In Poland, the results of health risk analysis only were the basis to prepare investment plan for water intakes, barely to improve treatment process of water.

In the risk analysis for groundwater intake, it is therefore necessary to carry out a fairly detailed inventory of objects and land use, hydrogeological conditions and the existing possible sources of groundwater pollution (Wardas *et al.* 2006). Such an inventory should include the recharge area of the intake and, in accordance with the guidelines in the Water Law Act, this area is designated by a 25-year isochrone. The risk analysis should include types of land

use and the potential scarcity of the resources of the abstracted water (qualitative aspect) and the capacity of the intake (quantitative aspect). In the recharge area of the intake, the existence of water facilities and locations of water use should be taken into account, i.e., discharge of sewage into waters or soil, including the use of sewage and the implementation of drainage devices or excavations as well as activities related to land drainage (building or mining drainage).

Risk analysis should include a comprehensive study on the location of industrial plants (mainly their type and impact on the environment, including mining plants, water intakes or others which are likely to significantly affect the environment), infrastructure (roads, railway lines, airports, car parks, campsites, car washes, cemeteries, residential buildings, and places of use and storage of chemicals for winter road maintenance), storage facilities (warehouses of petroleum products together with transmission installations), agricultural use (application of fertilizers and plant protection chemicals, grazing and watering animals, rearing or breeding fish, construction of silage prisms) and forest management – defining the type of forest crops (Koczera & Kalda 2016).

The above-mentioned facilities and methods of land use are inventoried on the basis of the existing hydrogeological, topographic and other thematic maps, depending on the specificity of the location of the water intake. However, the possible impact on the quality and quantity of water taken should be determined individually for a given facility. In these analyses, multi-annual data covering precipitation characteristics and data on water flows in nearby watercourses, which are hydraulically connected with the aquifer, within the range of the water runoff to the intake (Duda *et al.* 2013) are important. When determining the runoff area and designing the area of indirect protection, it is often advisable to use the results of isotopic studies of water age and tracer studies. Performing such tests is recommended for water intakes from deeper aquifers located in an area with complex hydrogeological conditions, inaccurately recognized water circulation routes and uncertain flow time from the recharge to the well of the intake, especially for a multi-hole covering a large area, as well as in the case of very large intakes. The methodology of performing

and interpreting the results of tracer tests is described in a guide edited by Zuber *et al.* (2007).

STUDY AREA

The groundwater intake 'Jarosław Dąbrowski' from a Carboniferous formation is located in the former shaft of the 'Sobieski' mine. This shaft was designed as a ventilation and filling system and was deepened in the years 1964–1965 using the rock mass freezing method. Due to high water inflows and inrush, the shaft had never been used for its primary function. Finally, in 1969, the shaft was sunk and transformed into a groundwater intake for drinking purposes. The intake is located in the southwestern part of the city of Jaworzno in the Upper Silesian Coal Basin. The exploitation water resources of the intake were determined as a result of geological works carried out in 1997 and they are $Q_e = 2,000 \text{ m}^3/\text{d} = 83.3 \text{ m}^3/\text{h}$ at the groundwater table at +189.68 m asl. The intake is a filter-free single deep-well with a water table in Carboniferous formations which are mainly developed as layers of dehydrated sandstones with small shale inserts and hard coal seams. The maximum range of the cone of depression, calculated for documentation purposes, is $R = 177.46 \text{ m}$. The recharge area is mainly made up of wasteland and urban areas, and is located in the old riverbed of the Przemsza River. The location and a simplified cross section are presented in Figure 1.

With regards to changes in the physicochemical parameters of groundwater from the Jarosław Dąbrowski intake, the hydraulic connection of the Upper Carboniferous aquifer and the Quaternary aquifer within the range of the Przemsza River valley is very important. The thickness of Quaternary layers in the intake profile is 26.7 m, and within the entire unit it is up to 30 m. These formations are permeable and remain in hydraulic connection with the lower Carboniferous formations below. The occurrence of periods of hydrological drought observed in the last decade, as well as low water levels in surface watercourses, adversely affect the chemical state of themselves and the associated water systems. The Jarosław Dąbrowski intake is located in an area of significant industrial and urban pressure. The recharge area is impacted by external factors,

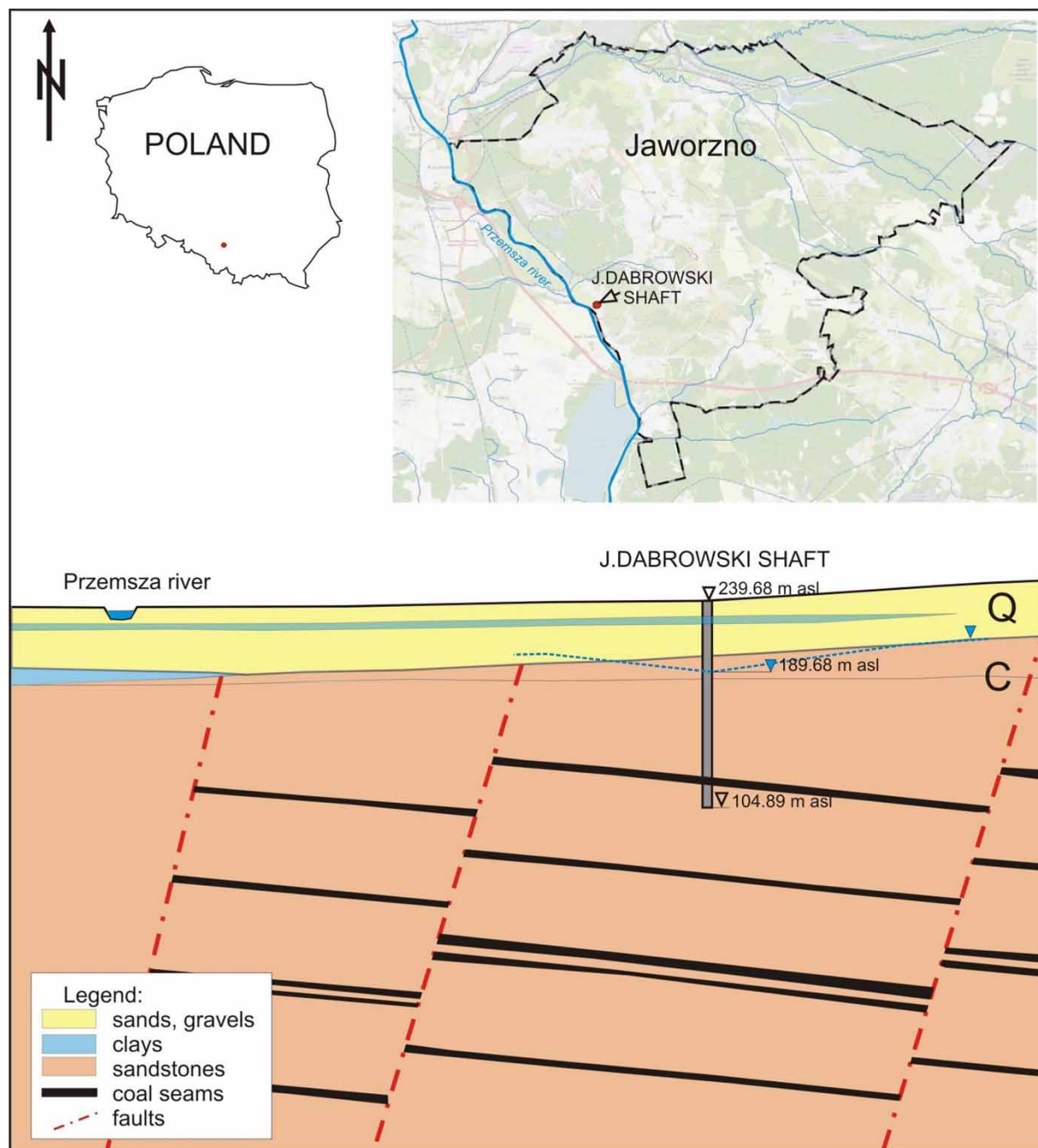


Figure 1 | Location of the Jarosław Dąbrowski shaft with a simplified hydrogeological cross section.

such as the discharge of industrial wastewater, mine waters, and areas of industrial waste landfills on the surface. The negative impact of hard coal mine dewatering is a particular threat to the quality of the groundwater in the study area, thus an increase in chloride content is observed.

As background for further investigations on health risk assessment methods and results, water quality monitoring data from May 2015 to May 2016 was collected. These data are collected by the operator of the intake – the Municipal Water Company in Jaworzno – and then published in

annual reports on the operation of water intakes (Municipal Water Company 2015/16). Water pH was in the range of 7.40–7.90 and the average annual pH value was 7.65. Throughout the year, the intake water temperature remained at 10.2 °C (minimum 6.20 °C to maximum 15.9 °C). Except for sporadic situations, in the analysed period the water from the intake was characterized by stability of the concentrations of such physicochemical parameters as iron, manganese, magnesium, calcium, ammonium ions and nitrite, and did not show significant fluctuations in the value of total hardness and alkalinity.

In the case of indicators characterizing the degree of water salinity, the annual concentration of chloride ions ranged from 232 mgCl⁻/L to 304 mgCl⁻/L and for sulfate ions 139 mgSO₄²⁻/L to 190 mgSO₄²⁻/L.

Water quality monitoring data revealing an upward trend in the content of chloride ions in water and analysis of the data of the location of the water table in the shaft during its operation from May 2015 to May 2016 in relation

to the monitored amount of chloride in raw water are presented in Figure 2.

The trend line for the time variation plot of the water table has been added to the chart to illustrate the general trend of decrease; however, only its illustrative nature should be taken into account, due to the continuous operation of the intake and the dynamic characteristics of the water table during pumping. Changes of chloride concentration in raw water from the intake are observed but the nature of the changes is complicated due to significant pressure of industry (underground coal mining area with continuous dewatering), specific geological and hydrogeological conditions with direct water inflow from the surface and from Carboniferous strata. For these waters increased high concentration of chlorides is characteristic. Due to significant drought derived from climate change, the upward trend of chloride content is observed, while water level in the intake, in general, remains stable.

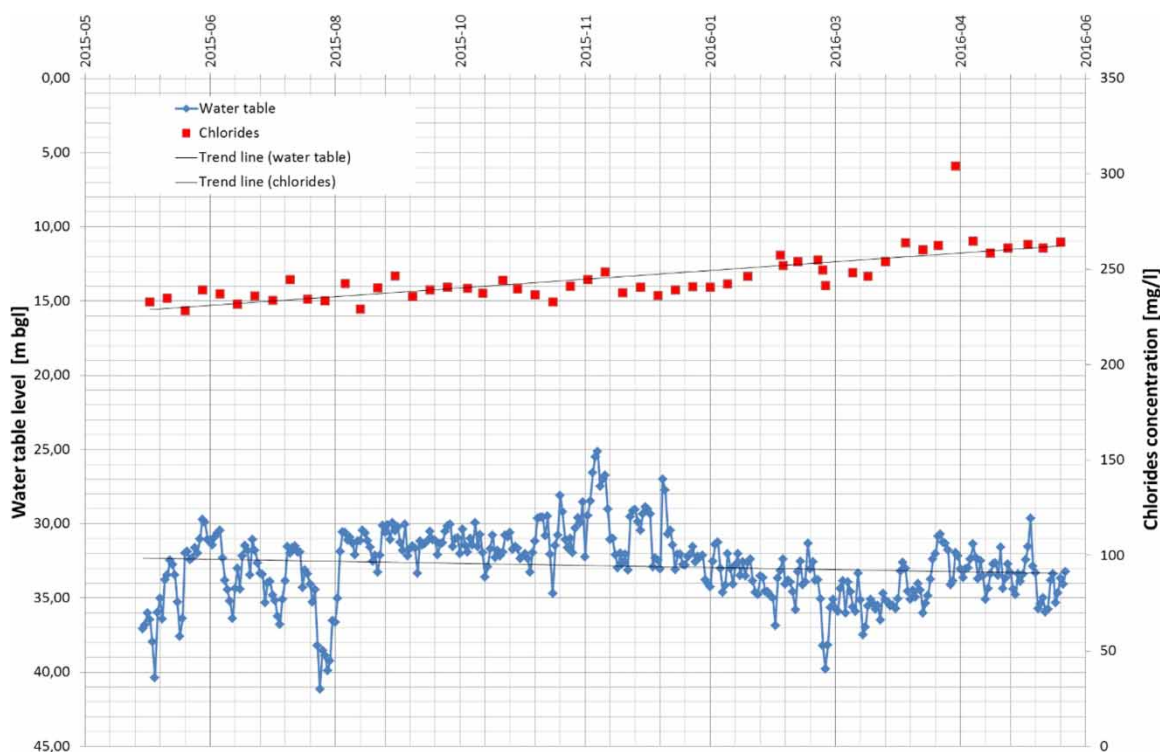


Figure 2 | Trends of chloride concentrations and water table in the Jarosław Dąbrowski intake in the period May 2015 to May 2016.

HEALTH RISK ANALYSES: METHODOLOGY

The determination of hazards for the water being used is important to define the factors that increase health risks due to the deterioration of its quality. Consuming low-quality water carries a risk of disease, such as cancer and diseases of the circulatory or digestive system. Highly mineralized inflows into the groundwater containing, among others, sodium (NaCl), potassium (KCl) and calcium (CaCl₂) salts, cause an increase in the chloride content, which in turn may lead to health risks for the consumers of these waters. In uncontaminated groundwater, the concentration of chlorides does not exceed 7–8 mgCl[−]/L. In waters contaminated with brine, the chloride concentration can be as high as 200–300 gCl[−]/L (Ziulkiewicz 2004). In the Regulation of the Ministry of Health of 7 December 2017 on the quality of water intended for consumption, it was determined that the concentration of chlorides in drinking water should not exceed 250 mg/L (taste threshold) due to the possibility of the deterioration of its organoleptic features.

Chlorides are components of extracellular fluid (88%) and affect the state of the body's water management. Increased salt supply may lead to hyperosmolality (the increase in the concentration of osmotically active substances in the extracellular fluid). Hyperosmolality causes increased secretion of vasopressin – a hormone that causes urine compaction by stimulating the resorption of water in the kidney tubules. Thanks to vasopressin, the volume of urine is reduced and the water resorbed in the kidneys is retained in the body. In addition, vasopressin affects vasoconstriction and increases blood pressure. Therefore, excessive salt intake is the cause of oedema, weight gain, the development of hypertensive disease, pulmonary oedema and cerebral oedema (Newsome 1979; Zelana 2012). Ingestion of excessive amounts of chlorides may also cause gastrointestinal symptoms (discomfort, mucous membrane damage and sometimes ulceration) (Makala 2016). Chlorides are not carcinogenic, but high sodium chloride intake may increase the carcinogenic potential of carcinogens, such as nitrosamines and the possibility of *Helicobacter pylori* stomach infection (European Food Safety Authority (EFSA) 2006; Gancz *et al.* 2008).

The World Health Organization (WHO) in the 'Guideline for drinking water quality' has determined the lethal

dose (*LD*₅₀) for rats (WHO 1996a). The data are collected in Table 1.

According to WHO recommendations, the daily intake of chlorides by adults should not exceed 5 g/d, and a dose of 0.6–3.6 kg/d is considered fully safe and has a non-harmful effect. The average daily intake of chlorides with food in the case of a salt-free diet is around 0.1 g/day (WHO 1996b). However, in Poland, the recommended dose of chloride is often exceeded by about three times, as the use of salt for cooking or in food processing increases the consumption of chlorides to as much as 12–15 g/day (Michalak-Majewska *et al.* 2013). Drinking water can also be a source of chlorides in the diet. Assuming that daily water intake is 2 L per person, the consumption of pure water with a low content of chlorides (up to 10 mg/L) is associated with the consumption of 20 mg of chloride per person, which is about 0.4% of the recommended daily dose of chlorides for an adult. However, if the concentration of chlorides in water is 250 mg/L, the daily consumption of chlorides with drinking water may amount to 0.5 g/L, which is 10% of the recommended daily dose of chlorides for an adult (Jarosz & Bułhak-Jachymczyk 2008).

Chlorides can also increase the corrosivity of water, and can react with equipment and the supply chain of water distribution. Chlorides increase the solubility of metals, resulting in an increase in their concentration in drinking water (Gil *et al.* 2018).

In order to analyse the health risk for the studied underground water intake in an old mine shaft located in a region of high pressure of mining plants on the water level, the following values of chloride concentrations were considered:

- *C*_{min} – 232 mgCl[−]/L
- *C*_{av} – 249 mgCl[−]/L
- *C*_{max} – 304 mgCl[−]/L.

The assessment of health risk and exposure was based on the recommendations of the US Environmental Protection Agency US EPA (US EPA 1989). In line with these recommendations, the risk assessment should first and

Table 1 | Fatal dose of chlorides for rats

<i>LD</i> ₅₀ (mg/kg)	CaCl ₂	NaCl	KCl
	1,000	3,000	2,430

foremost characterize the hazard, and then determine the value of the dose taken (Equation (1)), i.e., the amount of harmful substance that the body is exposed to in a given way of exposure per day per 1 kg of body weight:

$$I = \frac{C_w \cdot F_l \cdot K_w \cdot EF}{M_c} \quad (1)$$

where: I is the dose taken of the substance, the estimated daily intake in conditions of chronic exposure [$\text{mg} \cdot (\text{kg}^{-1} \text{d}^{-1})$]; C_w is concentration of a chemical substance in water [mg/L water]; K_w is magnitude of the exposure to a given environmental medium per unit of time [L water/d]; F_l is uncertainty factor – this number is defined within the range 0–1, which determines what part of the actual consumption comes from a contaminated source; EF is exposure factor; and MC is body weight (kg).

The exposure factor (EF) represents daily exposure to the contaminant. The EF is calculated by multiplying the exposure frequency by the exposure duration (ED) and dividing by the time period during which the dose is to be averaged (Equation (2)) (PHA 2005):

$$EF = \frac{F \cdot ED}{AT} \quad (2)$$

where: F is frequency of exposure and duration of exposure [days/year]; ED is exposure duration (years); AT is averaging time ($ED \times 365$ days/year).

The values used for the calculation are listed in Table 2.

The frequency of exposure was assumed by taking into account the number of days off work in a calendar year, which is approximately 114 days. It is assumed that we

spend half of this time away from home, so the frequency of exposure in a year is 308 days.

Subsequently, the health risk of chlorides supplied with drinking water was estimated by comparing the upper intake level (UL) with the value of the estimated daily intake (EDI) of chlorides with drinking water. UL is the dose of the substance that an adult person can safely be exposed to on a daily basis throughout life, without harmful impacts on health, according to current knowledge (Standing Committee on the Scientific Evaluation of Dietary Reference Intakes 2005). UL values for individual age groups are summarized in Table 3.

The daily intake (EDI) was estimated according to Equation (3):

$$EDI = F \cdot R \quad (3)$$

where: F is water consumption [L/person d^{-1}] and R is concentration of chlorides in water from intake [mg/L].

According to the recommendations of the US EPA, while it is impossible to accurately estimate the amount of water consumed from a given source, the largest possible value is estimated (US EPA 1989). The amounts of water consumed daily for a given age group and gender recommended by the Polish Experts Group (Woś *et al.* 2011) (Table 4) were used for calculations. In addition, it was assumed that 100% of drinking water is supplied from the intake.

In the last stage of calculations, the value of reference dose of chlorides in drinking water was compared with the actual collected dose by determining the hazard index (HI) (Equation (4)):

$$HI = \frac{I}{(RfD \cdot UF_{1-4} \cdot MF)} \quad (4)$$

Table 2 | Values of exposure parameters accepted for the calculation of the dose taken of the substance – I [$\text{mg}/(\text{kg d}^{-1})$]

Parameter of exposure	Children 1–3 years	Children 4–8 years	Children 9–12 years	Career women	Career men	Women retired	Men retired
C_k – Frequency of exposure and time of contact [days/year]	308	308	308	308	308	308	308
K_w – Range of contact with environmental medium [L water/d]	1.3	1.6	1.9	2.7	3.7	2.7	2.7
M_c – body weight [kg]	15	20	35	65	78	65	78
F_l – Part of daily intake from the source (with high level of hazard)	0.5	0.8	0.8	0.6	0.6	0.8	0.8

Table 3 | Upper intake level of chloride taken from various sources (*UL*) for different age groups

Age group	<i>UL</i> [mg/person d ⁻¹]
1–3 years	2,300
4–8 years	2,900
9–12 years	3,400
> 13 years	3,600

Table 4 | Recommended daily water consumption in Poland

Age group	Gender	Recommended daily water consumption [L]
1–3 years	Girls/Boys	1.3
4–8 years	Girls/Boys	1.6
9–13 years	Girls	1.9
	Boys	2.1
10–12 years	Girls	2.1
	Boys	2.4
13–15 years	Girls	2.2
	Boys	3
16–18 years	Girls	2.3
	Boys	3.3
19–70 years	Women	2.7
	Men	3.7
> 70 years	Women	2.7
> 70 years	Men	2.7
Pregnancy period	Women	3
Lactation period	Women	3.8

where: *HI* is hazard index; *I* is the dose taken of the substance (mg·kg⁻¹·d⁻¹); *Rfd* is reference dose (mg·kg⁻¹·d⁻¹); *UF*_{1–4} is uncertain factor; *MF* is modification factor (takes into account other uncertainties) (value 1–10); *UF*₁ takes into account species variability when extrapolating from animals to humans (value 10); *UF*₂ takes into account the individual variability within the human population (value 10); *UF*₃ is used in the extrapolation of data from the study of subchronic conditions to chronic conditions (value 10). In this study, it was assumed that the value of a *UF* is 100 (*UF*₁·*UF*₂·1) and *MF* is 1.

The *Rfd* is a benchmark dose operationally derived from the *NOAEL* or the *LOAEL* by consistent application

of generally order-of-magnitude uncertainty factors (*UF*s) that reflect various types of data sets used to estimate *RfDs*. The *Rfd* is determined by use of Equation (5):

$$RfD = \frac{NOAEL \text{ or } LOAEL}{UF_{1-4} \cdot MF} \quad (5)$$

The calculated value of *LOAEL* by EChA for NaCl was 2,533 mg·kg⁻¹·d⁻¹ (EChA 2020). It was assumed that if *HI* > 1, there is a possibility of negative health effects as a result of long-term exposure to water consumption with a defined (higher) chloride content. In the opposite case (*HI* < 1), the health risk is very low.

RESULTS AND DISCUSSION

Studies have shown that there is a relationship between chloride dose and the age and gender of people in terms of health risk assessment. In the case of boys and men, the consumption of chlorides with drinking water is much higher than in the case of girls and women of the same age. It was estimated that consumption of water with a chloride concentration in the range of 232–304 mg/L by boys aged 9–18 is associated with intake from 488 to 1,004 mgCl⁻/person d⁻¹, corresponding to 27 and 56% of the value of the upper tolerable daily consumption of chlorides with food (Figure 3). In this age group and for this gender, there is a risk of exceeding the upper intake level (*UL*), as a result of consuming water with an increased concentration of chlorides, which may result in the development of cardiovascular disease, the development of stomach cancer or the occurrence of a stroke. The amount of chloride taken together with drinking water increases with age. Men aged 19–70 are especially exposed to excessive supply of chlorides with drinking water. The estimated daily consumption of chlorides with water from the intake may be above 126 mg/person d⁻¹, which is 63% of the *UL* value. When leading an inadequate lifestyle and a chlorine-rich diet, there is a high risk of *UL* being exceeded in this age group and for this gender, which in turn, will be associated with the development of diseases.

Men at retirement age are less exposed to excessive chloride intake, due to the lower demand for drinking

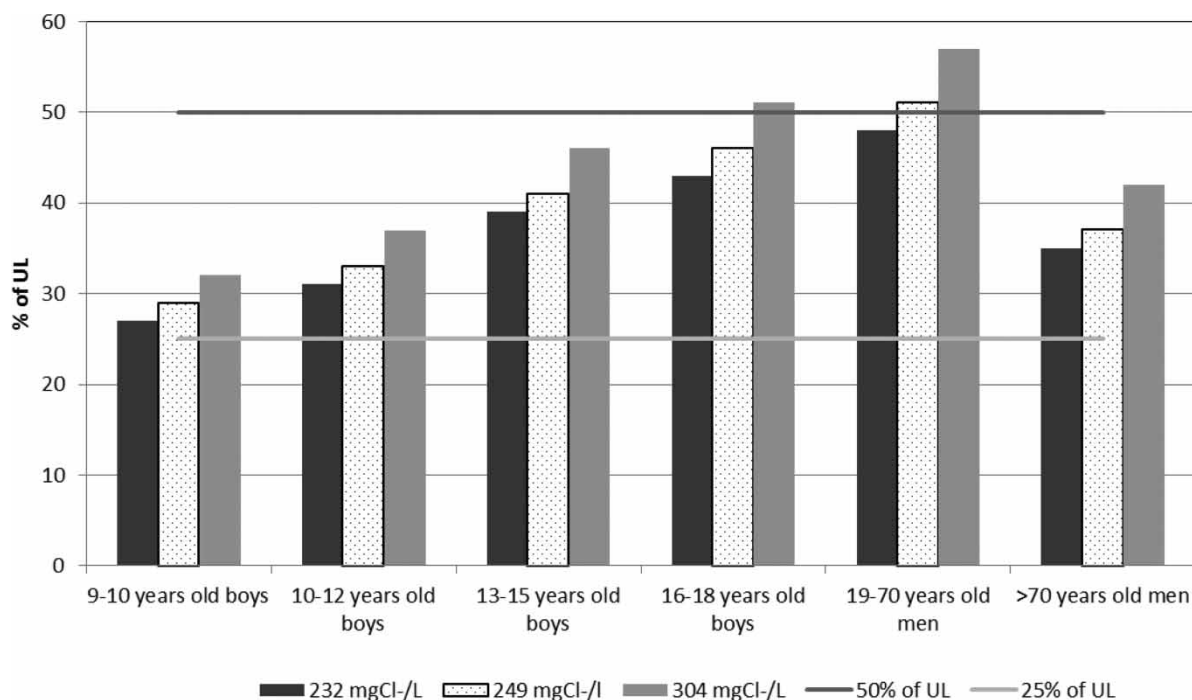


Figure 3 | Amount of chloride taken together with drinking water in relation to the *UL* value for the population of boys and men.

water in the diet. Consumption of water with a chloride content of 249 mg/L (value close to the allowable taste threshold of 250 mg/L) by a group of men aged 19–70 is associated with the consumption of half of the daily limit value of chloride. Consumption of water with chloride concentration above the taste threshold means that in the younger age group, i.e., boys aged 13–15, significantly increased chloride intake with drinking water can be observed (Figure 3).

In the case of girls and women, the amount of chloride taken with drinking water from the intake will be approximately 441 mg/L to 700 mg/L, which is 25 and 39% of the *UL* value, respectively. Pregnant women and women during lactation are among those women most exposed to excessive chloride intake. The consumption of chlorides with drinking water may amount to mg/person d⁻¹, which corresponds to 39–64% of the *UL* value (Figure 4).

Children aged 1–3 years and 4–8 years by drinking water with chlorides' concentration in the range of 301–486 mg/person d⁻¹ will consume 301–486 mg/person d⁻¹, representing 17–22% of the *UL* value (Figure 5).

In the last stage of the research, the value of the hazard index (*HI*) was determined. In the case of water with a chloride concentration of 232 mgCl⁻/L – a value below the acceptable threshold of 250 mgCl⁻/L, the highest value of the hazard index was observed for the group of children aged 1–12 years (*HI* was in the range 0.33–0.49). In the case of adults of working age, no adverse health effects are observed as a result of drinking water with a chloride content below the acceptable taste threshold (*HI* <0.25) (Figure 6).

The increase in chlorides' concentration in drinking water to 250 mgCl⁻/L is not a threat to people consuming this water (Figure 7).

Consumption of water with concentration of chlorides above the taste threshold level could pose a risk to children aged 4–8. The *HI* value for children was 0.64 (Figure 8).

It is important to note that risk assessment was calculated with the general assumption that water with increased chlorides' concentration covers total daily consumption, and the sensitivity analysis of this approach is required. The sensitivity analysis was performed on the assumption that both the chloride concentration in drinking

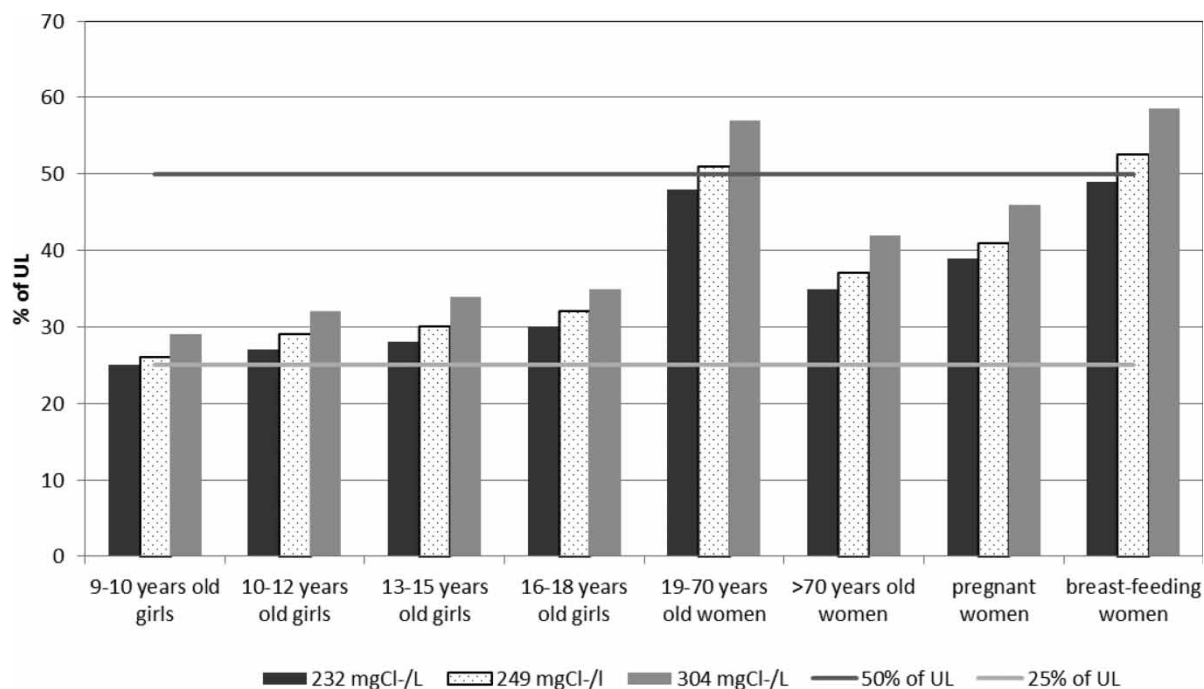


Figure 4 | Amount of chloride taken together with drinking water in relation to the UL value for the population of girls and women.

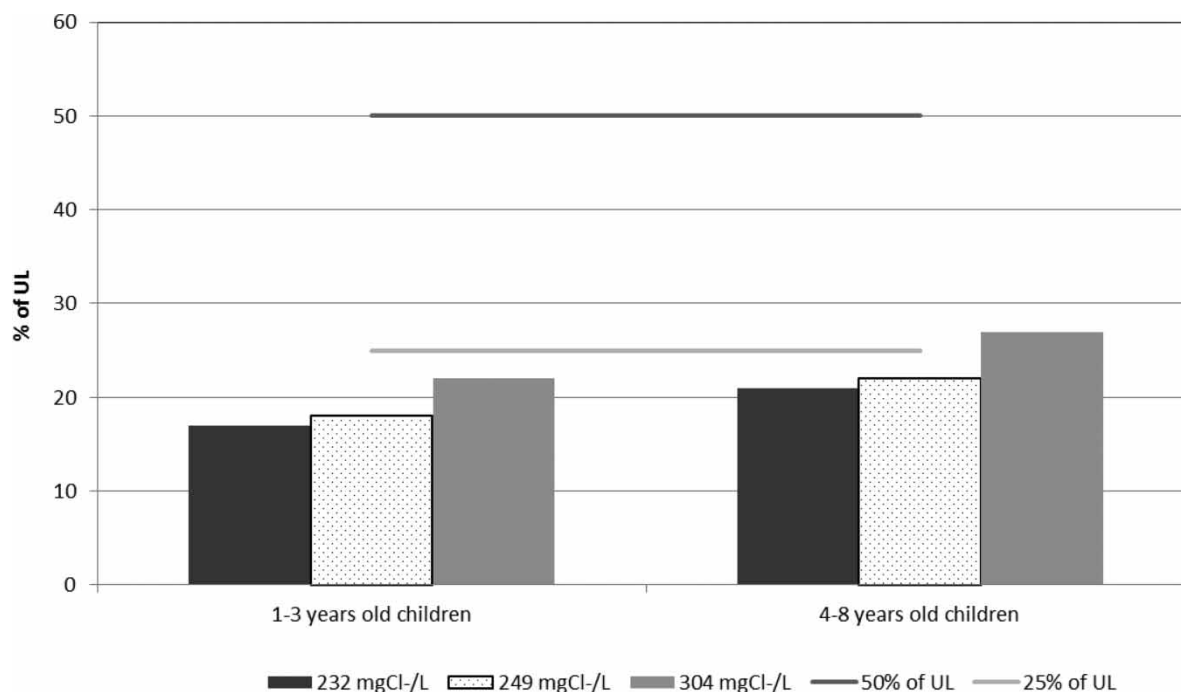


Figure 5 | Amount of chloride taken together with drinking water in relation to the UL value for the population of children aged 1–8 years.

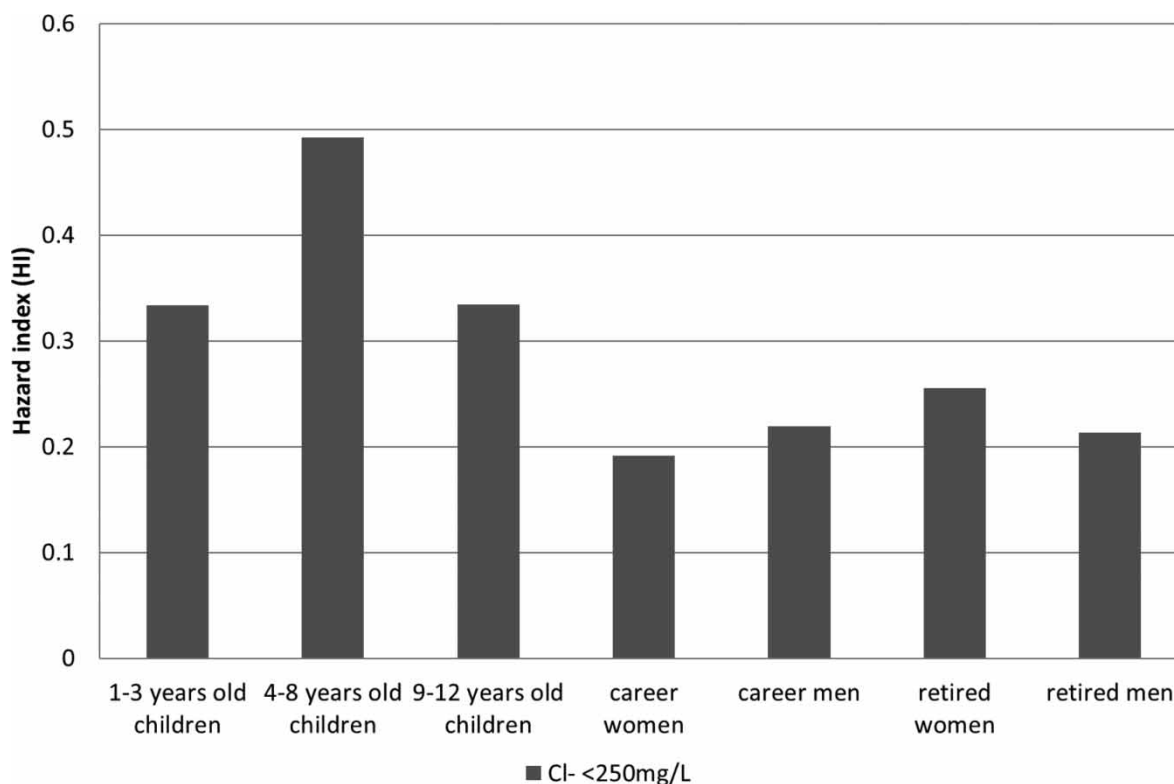


Figure 6 | Hazard index of potable water with chlorides' concentration below 250 mg/L for various gender and age groups of the general population.

water and the amount of water consumed from an analysed water intake affect the hazard index. The analysis was carried out assuming that the amount of water consumed from the water intake would be from 25 to 100% of the recommended daily intake. The sensitivity analysis was performed for the following chloride concentrations in water: 250 mgCl⁻/L, 300 mgCl⁻/L, 350 mgCl⁻/L, 400 mgCl⁻/L, 650 mgCl⁻/L. Concentration of 250 mgCl⁻/L in consumed water from concerned intake in the amount of 25–75% of K_w value will not pose a health threat to people. The high HI value (>0.8) was observed in the population of children of the age 1–8 when the K_w was 100% (Figure 9(a)). The concentration of 350 mgCl⁻/L poses a health hazard to children (1–8 years old) for whom the only source of water would be the water consumed from the intake (100% of K_w value). Children and the elderly will be not exposed to health effects when consuming less than 50% of the recommended daily intake. The high HI value (>0.8) was observed in the population of children in the age 1–12 and adults when the K_w was 75% (Figure 9(b)). Further increasing the concentration

of chlorides (≥ 350 mgCl⁻/L) could pose a health risk when consuming even 75% of the recommended daily intake (Figure 9(c) and 9(d)). The results of sensitivity analysis showed that $HI > 1$ will be observed for the adult population when the chloride concentration reaches the value 650 mg/L (Figure 9(e)).

CONCLUSIONS

In this paper, we presented an approach to health risk analysis for drinking water, in which the chloride concentration ranged between 232 and 304 mgCl⁻/L and was, on average, 249 mg/L. In the first step, the hydrogeological factors and relationship of chloride content in water from the old mine shaft were identified.

Conducting a broad environmental study for the needs of the risk analysis of a water intake is the first phase of work, which should then focus on analysing the quality parameters of raw water from the intake in order to track trends

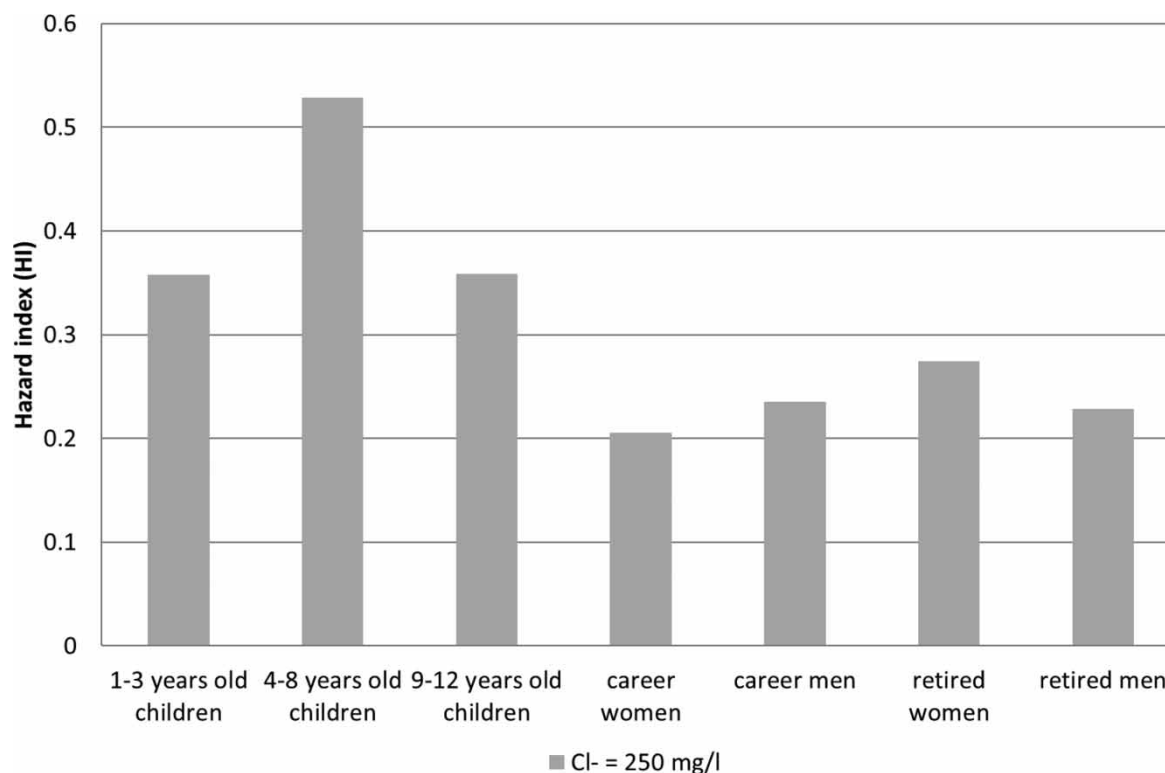


Figure 7 | Hazard index of potable water with chlorides' concentration equal to 250 mg/L for various gender and age groups of the general population.

in water chemistry changes, and then carry out health risk analysis.

Research revealed that the concentration of chlorides due to the impact of industrial, hydrogeological and mining factors would increase in the water from the intake. Health risk analysis is carried out as a part of legal regulations. In accordance with the requirements of WHO, in the first step, the value of the daily intake of chlorides per 1 kg body weight was determined. The calculations include the concentration of a chemical substance in a given environmental medium as well as the size, frequency and time of contact with a given medium. In the next phase, the calculated dose of collection was related to upper intake level value, i.e., to the dose of the substance that an adult person can consume daily throughout their life, without harmful impacts on health, according to the current state of knowledge. A relationship was found between chloride intake and the age and gender of those consuming the water. Men aged 19–70, in particular, are exposed to an oversupply of this mineral within their

drinking water. Daily consumption of chlorides with water from the intake in this group could account for up to 63% of the *UL* value. With an inadequate lifestyle and a high salt diet there is a high risk of *UL* being exceeded in this age group and for this gender, which in turn, was associated with the development of diseases. In the population of girls/women, pregnant women and women during lactation are those who are most exposed to excessive chloride intake in drinking water. The consumption of chlorides with drinking water in their case may amount to 39–64% of the *UL* value. The amount of chloride taken together with drinking water in the group of children aged 1–8 years was estimated at 17–22% of the *UL* value.

An important element of the risk analysis was the determination of the value of the hazard index. It has been found that children aged 1–8 years are most exposed to the negative effects of chlorides consumed with drinking water. In their case, the value of the hazard index was high even in relation to the consumption of waters with chloride concentration close to the permissible taste threshold. Older

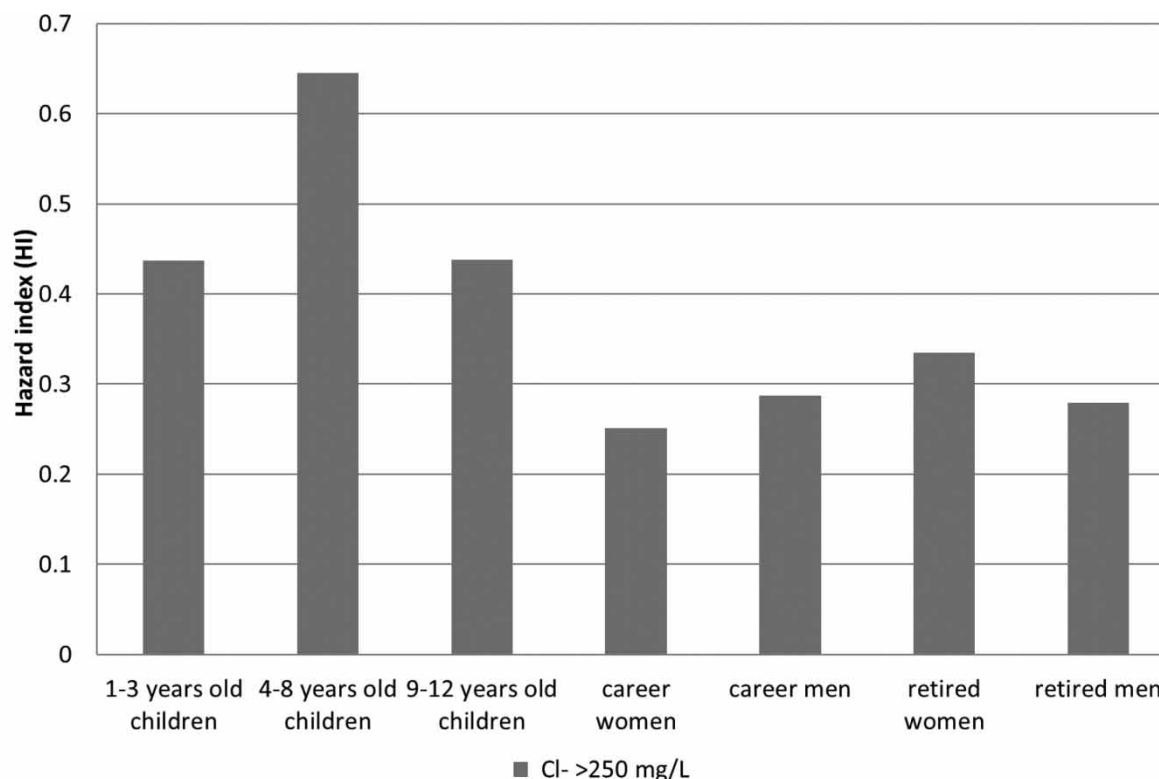


Figure 8 | Hazard index of potable water with chloride concentration higher than 250 mg/L for various gender and age groups of the general population.

children and adults are less exposed to the health effects caused by drinking water with chloride $\leq 250\text{--}300$ mg/L.

This research has shown how important it is, in the case of water intakes in highly urbanized and industrial areas, to recognize the factors that affect changes in water quality, which in turn, leads to the estimation of the health risk associated with the consumption of water from an intake with deteriorated quality. In the case of the unfavourable assessment for health risk, proposals should be made for actions in the resource area (protection zone) to limit the causes of negative changes in the quality of drinking water.

Studies have shown that there is a need to carry out a health risk analysis each time also for substances for which concentration monitoring is not required and there are no specific limit values. In the case of chloride, consuming water with a concentration close to the threshold value will have health effects for young children (1–8 years old). The methodology for determining the health risk presented in the paper and the results of the analysis have shown that it is easy to determine the health risk also for substances for

which the hazard parameters have not been clearly defined. The main objective of the study was to determine whether it is necessary to perform the health risk for substances for which concentration monitoring in raw water is not required and there are no threshold values, e.g., chlorides. The paper also presents an innovative approach to determining the health risk for non-toxic substances for which there are no clearly defined hazard parameters.

The results of the sensitivity analysis showed the importance of health risk assessment also for all non-carcinogenic substances in a concentration above the limit value. In addition, the results of study have shown that risk analyses should also be assessed for substances without a threshold value, when there is a suspicion of health hazards.

Due to the intake's location in a former coal field area, it is not possible to limit the impact of mining drainage and the introduction of saline waters into the environment for the protection of resources of abstracted drinking water. Its owner has no influence on external factors that are out of the protection zone of the considered intake – in this case,

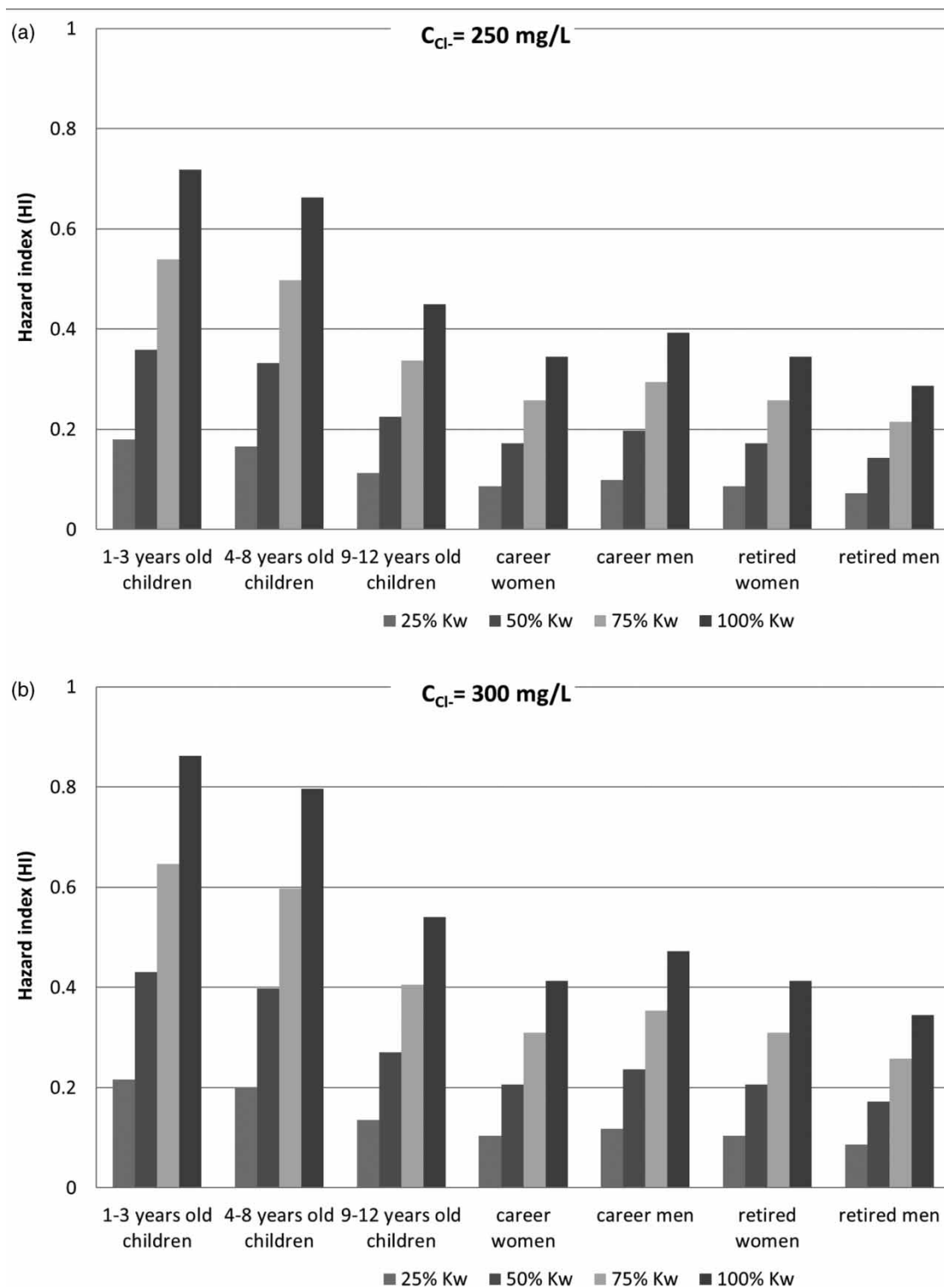


Figure 9 | The influence of the chlorides' concentration in the drinking water on the hazard index value for all age groups – sensitivity analysis (a)–(e) for various concentration values. (continued.).

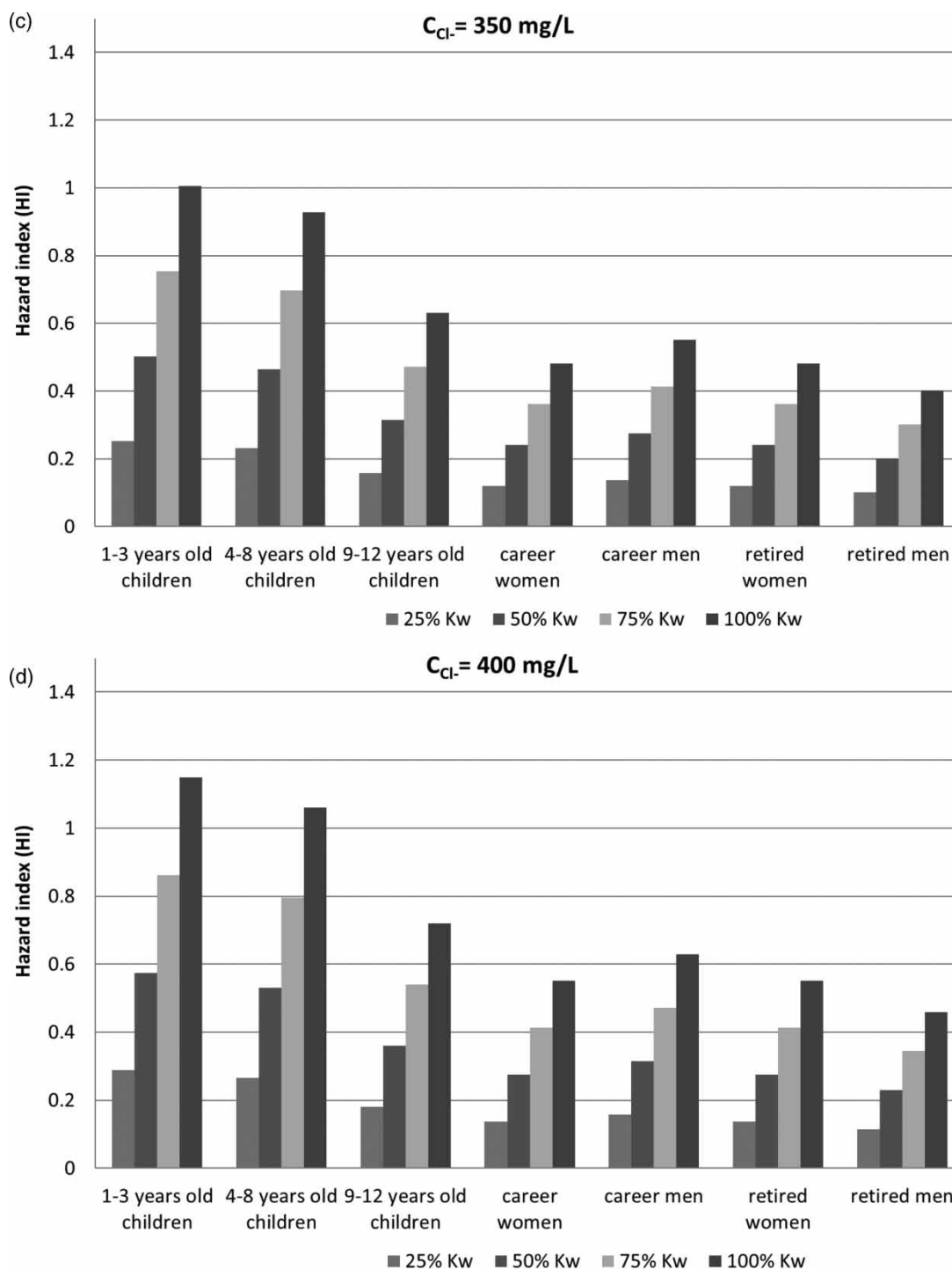


Figure 9 | (continued).

mining concessions, the need for drainage of the rock mass and industry impact on the water environment results in the application of technological solutions consisting of

equipping the water treatment plant located at the intake with a reverse osmosis plant installation, which ensures the reduction of chloride content in water to a level

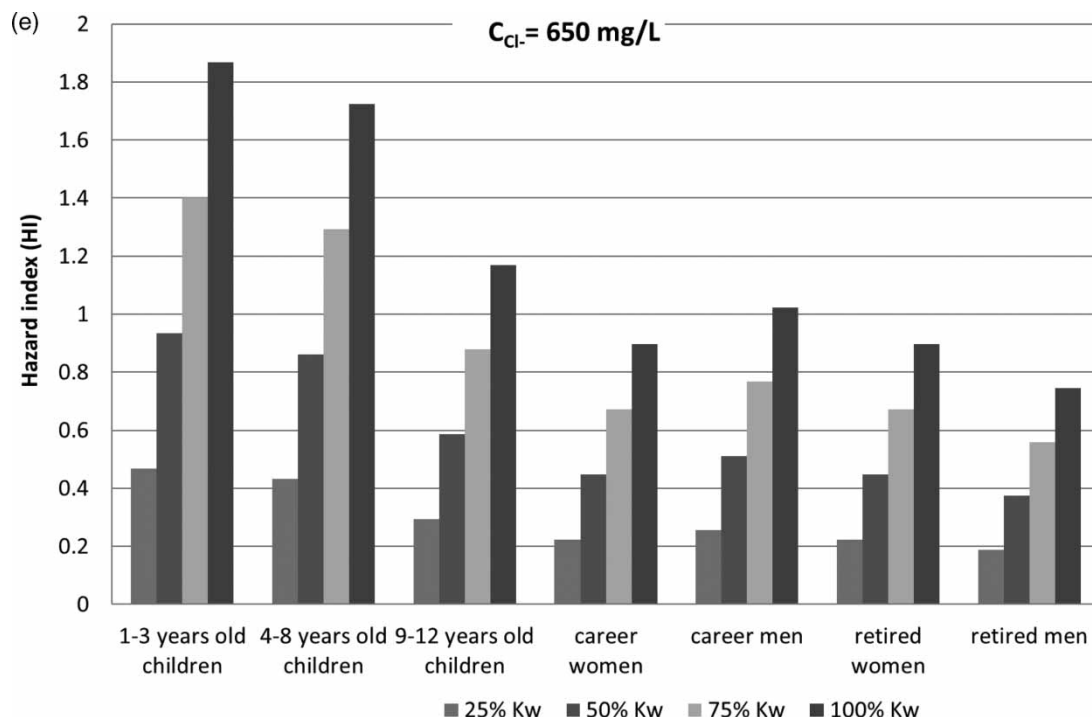


Figure 9 | (continued).

consistent with the requirements in this regard. The preliminary analysis of the health risk for water with an increased content of chloride ions carried out for the purpose of the intake was helpful in making the decision to implement the necessary technological solution – in this case reverse osmosis – in the water treatment plant. Risk analysis for groundwater intakes limited only to intake itself seem to be the first step to protect consumers against health risks and poor quality of delivered water.

DATA AVAILABILITY STATEMENT

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

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