

Article

# Water Quality Changes in Cement-Lined Water Pipe Networks

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Received: 12 February 2019; Accepted: 25 March 2019; Published: 30 March 2019



**Abstract:** This research focused on a model setup fed with underground water pumped into a water supply network. The scope of the research included a long- and a short-term stagnation of water in the setup and a water flow of 0.1 m/s. The water supplied into the system gradually lost its corrosive properties and developed calcium carbonate precipitation capability. These processes indicated that cement components migrate from the lining into the water. Apart from the primary components, the presence of the following trace elements was found: iron (399.6 ppb), magnesium (17.8 ppm), zinc (26.9 ppb), barium (22.6 ppb), boron (21.6 ppb), sodium (10.8 ppm), gallium (2.1 ppb). At early stages of the operation, recorded quantities were not significant. Leaching of the trace elements from cement increased after water was chlorinated. In turn, the highest percent increases were observed in the case of zinc (217%), boron (19%) and gallium (12%), whereas the increase for sodium, iron, barium and magnesium in water was insignificant (reaching a few percent). A repeated analysis of samples of stagnant water in the setup showed that most of trace elements were below the detectability level. Final concentrations of all elements identified in the water after its contact with internal cement mortar lining were much lower than the limit values set out for drinking water.

**Keywords:** water quality; internal cement mortar pipe lining; secondary water contamination

## 1. Introduction

Ductile iron is an alloy of iron and carbon in which, during solidification of the casting—owing to the addition of small amounts of magnesium or cerium—graphite nodules are produced. This form of graphite improves the mechanical properties of the material when compared to grey cast iron. Ductile iron pipes display higher plasticity, which makes them survive local strain without cracking and produces only local pipe deformations. These pipes are also highly resistant to lateral stress and high internal pressure. For the above reasons, for the past few decades they have been used for building water supply networks, especially in areas with shock risk, that is underneath motorways and streets with heavy traffic, in urbanised areas, in large urban and industrial agglomerations and in areas with complex geology. Ductile iron, just like most metal goods in contact with water, undergoes chemical and biological corrosion [1,2]. To prevent corrosion, different types of protective coatings are applied to the internal and external surfaces of the pipes. The most commonly used internal linings are made of cement mortar. In the case of the water intended for consumption, linings made of Portland cement or alumina cement are usually employed for contact with water characterized by greater corrosivity

(lower pH value, low alkalinity and high CO<sub>2</sub> content). External pipe surfaces are covered with zinc or zinc and aluminium coatings or with black varnish or epoxy plastic [3–5].

The primary ingredient of the cement is Portland clinker, its content—depending on cement type—ranging from 95% (additive-free Portland cement) to 15% (blast-furnace cement). Portland clinker is the product of heating (sintering) the mixture of limestone and aluminosilicate materials at the temperature of about 1450 °C. Primary elements contained in the cement include silicon, calcium, aluminium, iron and sulphur, possibly complemented with potassium, titanium, magnesium, manganese and phosphorus. During fuel combustion in rotary furnaces, some amount of heavy metals contained in the fuels are accumulated in the clinker. In recent years, different types of waste materials have been combusted in these furnaces, such as used motor oils or car tyres, which may have significantly higher heavy metal content than natural raw materials.

Elements contained in the primary components of the cement coating released into water in most cases do not affect its health claims. They do, however, have some impact on its organoleptic properties such as taste, odour or turbidity. Leaching of mechanical impurities, such as sand, may lead to corrosion of the fittings and any other components of water supply networks and systems [6–8]. Examination of damage to new cement coatings caused by water with varied corrosive properties conducted by Douglas and Merrill [9] showed that low-alkalinity aggressive water may significantly raise pH (from 7 to 12 after one week of testing), alkalinity and calcium content in water contacting cement mortar coatings. Application of a bitumen coating covering the cement layer considerably reduces the solubility of cement components [9]. Vik and Hedberg [10] listed the following water quality parameters that are recommended to be met in order to prevent dissolution of cement-based materials, including asbestos cement, cement pipes and cement coatings: pH > 7, alkalinity > 15 mg/L in the form of CaCO<sub>3</sub>, calcium >10 mg/L, sulphates < 200 mg/L, aggressive CO<sub>2</sub> < 5 mg/L.

Trace elements such as chromium, lead, zinc, nickel, arsenic, cadmium, vanadium or copper [11] are released from the cement lining into the water flowing through the pipeline. A study examining the leaching of trace elements from the cement, conducted by Achternbosch et al. [12] also showed that the use of waste materials for clinker firing increases element cement content to a minor extent. Reported relationships were not clear-cut however, as they were influenced by many factors, including method used and solubility testing. The study by van der Sloot [13] presented leaching of metals from cement mortar as a function of pH and duration of the period in which cement was in contact with water. Based on the revealed relationships, it was observed that the smallest amounts of metals leached into the water when it was neutral. Guo et al. [14] observed increased amounts of chromium, arsenic, barium and cadmium leached from the cement coating, recording their concentrations above the limit values for drinking water. Analyses conducted by Moudilu et al. revealed a correlation between leaching of heavy metals from the Portland cement and temperature, as well as the period of the experiment, using a new, highly sensitive method that allowed to determine very low concentrations of these elements [15]. Use of cement mortar coatings may lead to penetration of aluminium into the water, with the aluminium content depending on the type of cement. After two months of experiments on steel pipes with cement mortar lining, aluminium content was found to increase from 0.005 mg/dm<sup>3</sup> to 0.690 mg/dm<sup>3</sup>. This element affects human health and it is particularly dangerous for patients undergoing kidney dialysis [16]. Due to the fact that the leaching of aluminium from Portland cement occurs in low-alkalinity water, Berend et al. [16] recommend that internal coatings made in situ from regular Portland cement should not be used in water with an alkalinity lower than 55 mg/L as CaCO<sub>3</sub> and that factory-made internal cement coatings should not be used in water with alkalinity lower than 25 mg/L as CaCO<sub>3</sub>. It is also recommended that the use of high alumina cement lining should be limited in pipelines used for distributing drinking water, due to much higher aluminium content leached from this type of cement into the water, when compared to Portland cement [3,6].

The discussed topic is not new. However, previous studies focused on the laboratory, mainly static, leaching tests. This paper presents new studies conducted under real dynamic conditions, based on a new 90 m experimental set. The hydraulic conditions in this set reflected the average conditions in

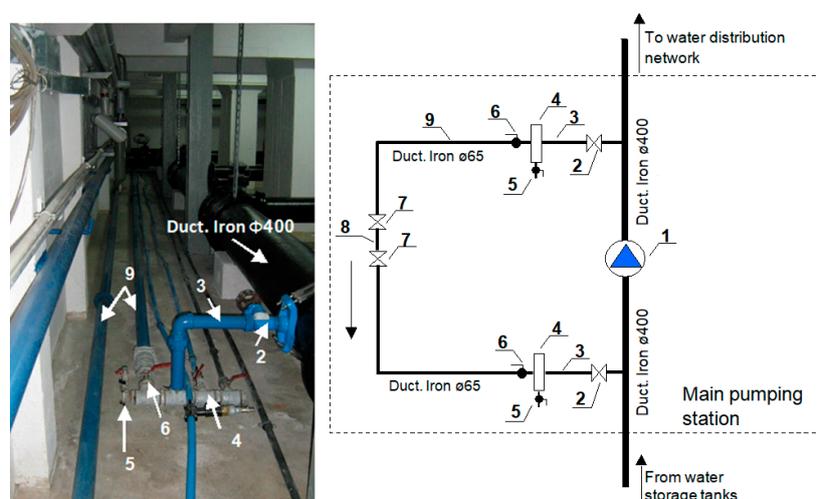
the entire network, determined on the basis of field measurements and numerical calculations (using a numerical model devised in EPANET 2.0 software, US EPA, Washington D.C., USA, 2000).

In this paper, the composition of cement lining was connected with the changes in the quality of water transmitted via lined pipeline. The occurrence of changes in the water quality was indicated both in short (hourly) and long-term (daily).

The available literature offers no exhaustive information on the impact of cement mortar lining of water conduits on the quality of the conveyed water. This is because the interaction between water and cement lining is determined by many factors; a significant role is played by water composition and type of lining but also properties of the contacting water. Also, state-of-the-art laboratory methods vary and quite often fail to reflect real-life conditions found in water supply networks. In light of the above, this paper presents a properly designed study conducted using the experimental setup, with the main purpose of determining the impact of cement coating covering the internal surface of a water conduit made of ductile iron on water quality in terms of changes in its physico-chemical properties.

## 2. Materials and Methods

The study was conducted in an experimental setup composed of the main pumping station supplying a water supply network in a selected city (Figure 1). Water was abstracted from intakes located in underground Cretaceous formations using 8 wells, from 55 to 70 metres deep. Abstracted water met the requirements for drinking water set out in WHO [17] and Polish National Standards [18] and except for periodic chlorination, it requires no treatment.



**Figure 1.** View (left) and schedule (right) of measuring stand; 1—a set of pump in pumping station II-nd degree, 2—valve  $\phi 50$ , 3—pipe, galvanized steel  $\phi 50$  mm,  $L = 1.2$  m, 4—splitter, a set of galvanized fittings  $\phi 50$ , 5—socket for sampling,  $\phi 15$ , 6—ball valve  $\phi 50$  mm, 7—valve  $\phi 65$ , 8—part of the ductile iron pipe for the microbiological testing  $\phi 65$ , 9—ductile iron pipes  $\phi 65$  lined with cement mortar,  $L = 80$  m.

The experimental setup was made of ductile iron pipes with a total length of 80 m and an internal diameter of 65 mm. The working volume of the experimental setup was  $250 \text{ dm}^3$ .

The study was composed of three testing series:

Series I—long-term (multiple day) stagnation of water in the setup. Duration of the series until stabilisation of water quality in the setup. Water samples were collected once a week.

Series II—short-term (few hour) stagnation of water in the setup. Water samples were collected from the setup after 0, 1, 2, 4, 8, 16 and 32 h.

Series III—water flow through the installation at the velocity of 0.1 m/s. Water samples for the analyses were collected during the supply and drainage of the water from the setup, considering retention times of the water in the pipeline.

The experimental setup was chlorinated and washed before every series. Before each series of measurements, a water sample was collected in order to determine the initial conditions. Physico-chemical analyses of water samples determining changes in its quality were conducted in accordance with applicable analytical standard methods. The standards according to which the measurements of particular water quality parameters were conducted are presented in Table 1. The analysis of every water quality parameter in each sample was performed in triplicate and the average of the results are presented in the tables. Trace element content in the water and in the cement coating was measured using the JY238 Ultrace Inductively Coupled Plasma—Optical Emission Spectrometer (ICP-OES) (Jobin Yvon-Horriba, Montpellier, France) [19]. The calibration solutions were prepared using CentiPUR<sup>®</sup> VIII multi-element calibration solution (Merck group, Darmstadt, Germany, 2013). In order to conduct a more accurate quantitative determination of elements, the investigations were carried out in two phases. In the initial phase, a fast semi-quantitative analytic methodology was employed. This phase, as well as the previous results of studies on cement lining, enabled the determination of the elements that had to be subjected to further quantitative indication with a more accurate method (second phase). In that phase, determination was carried out by means of inductively coupled plasma mass spectrometer (ICP-MS, 7700x, Agilent, Palo Alto, USA). Similar to the initial phase, the quantitative determinations were performed using calibration curves and the same pattern. The scope of analytical control of water samples is presented in the tables containing results of relevant analyses.

**Table 1.** Quality of the water feeding the experimental set-up.

Parameter	Unit	Sample					Limit Value for Drinking Water [17,18]	Standard Method
		No 1	No 2	No 3	No 4	No 5		
temperature	°C	10.5	10.8	12.4	11.0	10.2	-	-
colour	mg Pt/L	1	1	1	1	1	15	PN EN ISO 7887:2012
turbidity	NTU	0.48	0.45	1.04	0.89	0.55	1	PN EN ISO 7027:2003
pH	-	7.06	7.18	6.90	7.53	7.11	6.5 ÷ 9.5	PN EN ISO 100523:2012
alkalinity	mval/L	5.9	5.7	5.4	6.3	6.2	-	PN EN ISO 9963-1:2001
hardness	mval/L	7.4	7.3	6.8	7.8	8.2	1.2 ÷ 10	PN ISO 6059:1999
calcium	mg Ca/L	122	119	113	104	122	-	EDTA Method
iron	mg Fe/L	0.08	0.07	0.24	0.10	0.07	0.2	PC Multidirect Method 220
manganese	mg Mn/L	0.018	0.015	0.021	nd	nd	0.05	HACH Method 8149
chlorides	mg Cl/L	17.8	18.2	21.3	32.0	39.0	250	PN ISO 9297:1994
sulphates	mg SO <sub>4</sub> /L	72	71	60	86	82	250	HACH Method 8051
chemical oxygen demand	mg O <sub>2</sub> /L	1.2	1.3	4.6	1.8	0.8	5	PN-74/ C-04578-03
ammonium ion	mg H <sub>4</sub> /L	0.005	0.008	0.013	0.090	0.141	0.5	HACH Method 8038
nitrites	mg O <sub>3</sub> /L	11.3	9.7	12.0	15.8	9.2	50	HACH Method 8039
free CO <sub>2</sub>	mg/L	9.2	9.4	50.6	50.6	35.2	-	PN -74/ C-04547/01
aggressive CO <sub>2</sub>	mg/L	nd	nd	nd	nd	nd	-	PN -74/ C-04547/03
oxygen	mg O <sub>2</sub> /L	7.8	8.1	7.7	6.7	5.3	-	Winkler Method
dry residue	mg/L	389	358	542	494	521	-	PN -75/ C-04616/01

### 3. Results

#### 3.1. Analysis of Test Results for Water Feeding the Setup, Considering Its Corrosive Properties

Based on the obtained results of the physico-chemical analysis (Table 1), it may be found that the setup for the entire period of the experiment was fed with water of variable quality due to employment of different wells in the intake. Changes in carbon dioxide and dry residue concentrations, that is, parameters significant in terms of corrosive properties of the water, are particularly clear. In general, feeding water had a pH approximating neutral (pH ranging from 6.90 to 7.53), very low colour (1 mg/L) and turbidity (from 0.45 to 0.89 NTU, single result of 1.04 NTU), as well as moderate salinity, which is primarily reflected by the values of such indicators, as general hardness (6.8 ÷ 8.2 mval/L), alkalinity (5.4 ÷ 6.3 mval/L), chloride (17.8 ÷ 39.0 mg Cl/L) and sulphate content (60 ÷ 86 mg SO<sub>4</sub>/L), as well

as dry residue content (358 ÷ 542 mg/L). Iron concentration failed to exceed 0.10 mg/L (single result of 0.24 mg/L was reported), whereas manganese concentration did not exceed 0.021 mg/L. Organic matter burden may be regarded as relatively low due to the values of chemical oxygen demand (0.8 ÷ 1.8 mg/L; single value of 4.6 mg/L), ammonium ion (0.005 ÷ 0.141 mg/L) and nitrate ion (9.2 ÷ 15.8 mg/L) indicators. The analysed water was reported to contain gas admixtures, such as carbon dioxide with concentration ranging from 9.2 to 50.6 mg/L and oxygen (5.3 ÷ 8.1 mg/L).

When comparing the quality of the feeding water with standards set out for water for human consumption [17,18], it should be stated that in terms of compliance with these standards, the water quality raised no concerns as regards the analysed physico-chemical indicators.

The basic indicators used to approximately identify water impact on the cement coating are the Ryznar Stability Index (RSI) and the Langelier Saturation Index (LSI), as well as an index proposed by American Water Works Association (AWWA) [20]. Additionally, some indicators referring to specific water quality parameters may be determined, allowing evaluation of their impact on cement mortar lining. In order to provide a complete picture of the properties of water supplying the experimental setup in terms of its chemical stability and corrosive properties in reference to typical metals used in constructing water distribution networks and systems, indices proposed by the technical standards [19–24] regarding impact of water on metallic materials were examined.

Based on the analysis of the indices determining the chemical stability of the water (Table 2), it can be concluded that, although water quality varied, it posed a minor hazard for cement and metallic materials. Only single results we obtained indicate some risk of corrosion. The only exception is the Ryznar Stability Index, with values always within the range typical for aggressive water.

**Table 2.** Corrosion indicators of the water feeding the experimental set-up.

Parameter	Sample					Small Risk of Corrosion, When	
	No 1	No 2	No 3	No 4	No 5		
Langelier index ( $I_L$ )	−0.04	0.08	−0.40	0.23	−0.09	0	
Ryznar stability index ( $I_R$ )	7.14	7.02	7.70	7.07	7.29	6.25 ÷ 6.75	
AWWA index ( $I_{AWWA}$ )	12.1	12.2	11.9	12.5	12.2	>12	
corrosion indices for metallic materials							
Copper	S	8.1	8.1	9.0	5.8	7.6	>1.5
zinc-coated	$S_1$	0.4	0.4	0.4	0.6	0.5	<0.5
ironic materials	$S_2$	11.0	12.7	9.6	10.6	18.2	>3
corrosion-resistant steels	$c(Cl^-)$	0.5	0.5	0.6	0.9	1.1	<1.5 mmol/L
	$c(O_2)$	7.8	8.1	7.7	6.7	5.3	>3 mg/L
	pH	7.1	7.2	6.9	7.5	7.1	>7
Cast iron	$c(HCO_3^-)$	6.1	6.0	5.7	5.2	6.1	>2 mmol/L
	$c(Ca^{2+})$	3.1	3.0	2.8	2.6	3.1	>1 mmol/L

$I_L = pH - pH_n$ ,  $I_R = 2pH_n - pH$ ,  $I_{AWWA} = pH + \log(AH)$  where: pH—real,  $pH_n$ —saturation, A—alkalinity [mg  $CaCO_3/L$ ]; H—calcium carbonate hardness [mg  $CaCO_3/L$ ],  $c(HCO_3^-)$ —bicarbonate ion concentration [mmol/L];  $c(SO_4^{2-})$ —sulphate ion concentration [mmol/L];  $c(Cl^-)$ —chloride ion concentration [mmol/L];  $c(NO_3^-)$ —nitrate ion concentration [mmol/L]. S,  $S_1$ , and  $S_2$ —calculated using following formulas:

$$S = \frac{c(HCO_3^-)}{c(SO_4^{2-})}, \quad S_1 = \frac{c(Cl^-) + c(NO_3^-) + 2c(SO_4^{2-})}{c(HCO_3^-)}, \quad S_2 = \frac{c(Cl^-) + 2c(SO_4^{2-})}{c(NO_3^-)} \quad (1)$$

### 3.2. Analysis of Test Results for Multiple Day Stagnation of Water in The Setup

The first series assumed stagnation of water in the setup until stabilisation of its properties. In this series, water samples were collected once a week for 8 consecutive weeks. Changes in the values of the most significant water quality indicators over the experimental period presented in Table 3 provides values of the basic water stability indices in subsequent weeks of stagnation.

**Table 3.** Corrosive properties of the water during many weeks' stagnation in the experimental set-up (series I).

Week	I <sub>L</sub>	I <sub>R</sub>	I <sub>AWWA</sub>
0	−0.04	7.14	12.09
1	0.08	7.02	12.20
2	0.31	7.04	12.67
4	0.54	6.71	12.89
5	0.81	6.49	13.13
6	0.81	6.64	13.21
7	0.92	6.48	13.39
8	0.76	6.74	13.15
water stable for	0	6.25 ÷ 6.75	>12

Obtained results show that over the stagnation period, water reacted with the cement coating for the first 5 weeks. Over this period, a gradual increase in pH was observed (from 7.06 to 8.11), which was accompanied by carbon dioxide disappearance and increased water turbidity, chloride content and in particular sulphate and dry residue content. During this period, alkalinity and water hardness was reported to increase to a lesser extent. Values of the Langelier Saturation Index (LSI) indicated that water introduced into the setup gradually lost its corrosive properties and the calcium-carbonate balance was observed. The above-mentioned changes in water quality reflected gradual saturation of the water with cement coating components and an increasing water capability to precipitate calcium carbonate deposits. Analyses of water stagnant in the setup over the subsequent period of the research confirmed this trend, which is especially emphasized by the reduced value of water turbidity, calcium concentration and dry residue content. It should be emphasized that water quality over the entire experimental period corresponded to the quality of water for human consumption, whereas values of the AWWA index showed that water could be safely used in contact with cement materials.

### 3.3. Analysis of Test Results Obtained Over a Few-Hour Stagnation of Water in the Setup

The second series involved examination of water stagnant in the setup at time intervals of 1, 2, 4, 8, 16 and 32 h. The sample used consisted of water found in the setup after about 11 months of its operation. The tests were intended to show changes in water quality over a short-term contact of water (used in the water supply practice) with the cement coating covering the internal surface of the conduits. Results of physico-chemical analyses of water samples are presented in Table 4.

**Table 4.** Corrosive properties of the water during stagnation period in the experimental set-up (series II).

Hour	I <sub>L</sub>	I <sub>R</sub>	I <sub>AWWA</sub>
0	0.33	6.87	12.64
1	0.32	6.88	12.64
2	0.41	6.69	12.56
4	0.47	6.63	12.64
8	0.48	6.62	12.68
16	0.48	6.62	12.71
32	0.56	6.49	12.73
water stable for	0	6.25 ÷ 6.75	>12

Analyses of individual water samples show changes in water quality after prolonged contact with cement-mortar lining. The rate of these changes was very slow and they were related to penetration of cement components found in the coating into the water; this mainly concerned calcium (aluminium content remained very low and rather stable). Reactions between water and cement led to increased turbidity, pH and gradual reduction of carbon dioxide content. As far as provisions concerning water intended for human consumption are concerned, water turbidity had excessively high values.

Changes in the physico-chemical indicators of water quality were accompanied by changes in the values of stability indices. The increased Langelier Saturation Index and decreased Ryznar Stability Index reflected increased water capacity to precipitate calcium carbonate, whereas increased AWWA index—safe use of the applied coating in case of contact with the examined water.

#### 3.4. Analysis of Test Results for Water Flowing through the Setup at the Velocity of 0.1 m/s

The third series was conducted under dynamic conditions, that is, water flowing through the experimental setup at a velocity of 0.1 m/s. This rate is commonly used in the currently over-dimensioned water supply networks. Water samples were collected after 11 months of using the setup. During sampling, a 13-min long contact of water with the cement-mortar lining was considered. Results of the analysis for this series are provided in Table 5.

**Table 5.** Quality of the water flowing through the experimental set-up at the velocity of 0.1 m/s (series III).

Parameter	Unit	Water Feeding the Setup	Water Flowing Out from the Setup
temperature	°C	11.0	11.2
colour	mg Pt/L	1	1
turbidity	NTU	0.89	0.93
pH	-	7.53	7.58
alkalinity	mval/L	6.3	6.4
hardness	mval/L	7.8	8.2
calcium	mg Ca/L	104	114
iron	mg Fe/L	0.10	0.055
manganese	mg Mn/L	0	0
chlorides	mg Cl/L	32.0	39.0
sulphates	mg SO <sub>4</sub> /L	86	86
oxygen consumption	mg O <sub>2</sub> /L	1.84	1.12
ammonium ion	mg NH <sub>4</sub> /L	0.09	0.09
nitrates	mg NO <sub>3</sub> /L	15.8	15.8
free CO <sub>2</sub>	mg/L	50.6	39.6
oxygen	mg O <sub>2</sub> /L	6.70	6.93
dry residue	mg/L	494	519
Langelier saturation index (IL)	-	0.23	0.38
Ryznar stability index (IR)	-	7.07	6.82
AWWA index	-	12.64	12.67

A comparison between the quality of water feeding the setup and water flowing out from the setup shows that even after such short-term contact of water with the cement-mortar lining, under flow-through conditions there occurred an interaction between the water and the cement mortar lining. It should be stressed that changes in water quality proved minor, however they have already provided evidence of penetration of the basic cement components into the water. This increased the values of basic water quality indicators, including in particular turbidity, pH, hardness and dry residue content and decreased carbon dioxide water concentration. Values of indicators determining water stability point out that during contact with cement mortar lining, the water increased its capability to precipitate calcium carbonate.

#### 3.5. Determination of Trace Element Content in Water Contacting Cement-Mortar Coating

On the basis of the results of the analysis of a sample cement coating covering the interior surface of the pipeline in the experimental setup using the Inductively Coupled Plasma—Optical Emission Spectrometer (ICP-OES), it was found that, besides the primary cement components (including such elements as potassium, sodium, magnesium, calcium, zinc, aluminium) it also contained numerous trace elements such as iron, manganese, barium, chromium, lead, nickel and cobalt. Analyses of

chlorinated and non-chlorinated water contacting the cement-mortar coating conducted at the initial stage of setup operation (preliminary research) allowed to make a general statement that majority of trace elements found in the cement were successfully immobilized and significant amounts failed to penetrate the water that contacted the coating. It should be stressed however, that leaching of trace elements from the cement was less intense in the case of chlorinated water [25].

As part of the research presented herein, an attempt was made to analyse the phenomenon of trace element penetration into the water after long-term use of the setup. To that end, after eight months of using the setup, another test for trace element water content was performed. The analysed sample consisted of water feeding the setup and water stagnant in the setup for 32 h. The results are presented in Table 6. The majority of trace elements (including those detectable in the water at the initial stage of setup operation) were below the detection limit of the spectrometer. As regards elements detected in the stagnant water on the other hand, there was an insignificant increase in their concentration when compared to the water feeding the experimental setup, which was reported for sodium, magnesium, iron and barium. A slightly larger increase of about 19% was reported only in the case of boron concentration whereas there was a slightly lower increase of about 12% in the case of gallium. A large (three-fold) increase was reported for zinc on the other hand, although its final concentration in the stagnant water (0.027 mg/L) is still found to be rather low. It should be stressed that zinc content in drinking water is not limited by any Polish, American or international provisions. All final concentrations of elements detected in the water after its contact with the coating were considerably below limit values for drinking water set out in relevant legal provisions.

The reported small increases in their concentration is likely to occur for a number of reasons. The first one is the effective immobilisation of trace elements in the structure of the cement (their presence is confirmed, therefore their possible low content should be excluded). There are studies [26,27] that proved that well-maintained cement provides for small leaching of hazardous elements, even if it contains an admixture of ashes obtained from incinerated hospital waste. The question of composition and maintenance of the cement intended to contact drinking water therefore requires special attention. The second reason for a minor increase in metal water concentration may be the fact that the examined cement-mortar lining was in permanent contact with the water for a long time (8 months), therefore trace elements contained in the cement had already been leached earlier (this is suggested by preliminary research [25]). This brings about the need not only to maintain the mortar in water conduits but also to properly wash the conduits at the initial stage of use.

**Table 6.** Concentrations of trace elements in the water after 32-h stagnation in the cement-lined pipe (after 8 months in service).

Element	Unit	Water Feeding the Setup	Water after 32 h of Stagnation	Increment	Increment (%)	Limit Value for Drinking Water [17,18]
B	ppb	18.1	21.6	3.5	19	1000
Na	ppm	9.9	10.8	0.9	9	-
Mg	ppm	17.7	17.8	0.1	1	125
Fe	ppb	366.4	399.6	33.2	9	200
Zn	ppb	8.5	26.9	18.4	217	-
Ga	ppb	1.9	2.1	0.2	12	-
Ba	ppb	21.7	22.6	0.9	4	700

#### 4. Summary

The aim of this research was to examine the impact of cement-mortar lining covering the interior surfaces of ductile iron water conduits on the quality of water conveyed in these conduits. Tests were performed using an experimental setup made of commercially available pipes. The setup was supplied with underground water collected and provided, without treatment, to a water supply network in a mid-sized city. Water examined during the experiment was of variable quality, however for the

entire period of the experiment it displayed good physico-chemical properties in terms of its fitness for consumption, as well as poor corrosive properties with a tendency to form protective coatings, which made it a minor hazard for cement and metallic materials.

During the experiment, attempts were made to determine changes in the physico-chemical quality of the water during long-term (multiple day) and short-term (few hours) stagnation of the water in the setup and during its flow through the setup at the velocity of 0.1 m/s (which is typical of working water supply networks). While examining the impact of cement mortar lining on water quality, changes in the quality in terms of increased indicators such as primarily turbidity, pH, alkalinity, hardness, chloride, sulphate and dry residue content were reported, as well as reduced carbon dioxide water content. Values of indices characterising water stability pointed out that water introduced into the setup gradually lost its corrosive properties and acquired an increasing ability to precipitate calcium carbonate. This provided evidence of the penetration of components of the cement lining into the water. Such changes were recorded incrementally for the first 5 weeks of water contacting the cement lining. After shorter contact (up to 32 h), water quality changes were primarily related to increased turbidity and hardness and minor pH rise. The phenomenon of interaction between water and cement-mortar lining was also observed while testing the water flow through the setup at the velocity of 0.1 m/s. In this case, the change in water quality was very small. It did confirm, however, the leaching of cement components into the water, which raised the water's ability to precipitate calcium carbonate.

In the experiment, besides changes in the typical water quality indicators, an attempt was made to determine water penetration by metals found in the cement-mortar lining due to the applied cement manufacturing method. While testing the sample collected from the cement-mortar lining, besides the primary cement components, the following trace elements were detected: iron, magnesium, manganese, zinc, barium, chromium, lead, nickel and cobalt. Analyses conducted of samples of chlorinated and non-chlorinated water having contact with the cement lining at the initial stage of setup operation failed to confirm penetration of significant amounts of trace elements into the water (whereas leaching out of trace elements from cement was more intense in chlorinated water). Repeated analyses of samples of water stagnant in the setup for 32 h, after 8 months of setup operation, showed that the majority of trace elements, including those detected at the initial stage of operation, were below the detection limit. Among elements detected in the stagnant water, the largest increase in content with respect to the feeding water was observed for zinc, boron and gallium. Increases in sodium, iron, barium and magnesium water content after stagnation were scarce. It should be stressed that final concentrations of all elements detected in the water after its contact with the cement-mortar lining were much lower than the permitted limits for drinking water. Low concentrations and small (excluding zinc) increases in element concentrations were most probably caused by their effective immobilisation in the cement structure and by earlier leaching from the lining.

**Author Contributions:** Conceptualization, methodology, writing—original draft preparation, J.W., M.K.; investigation, resources, D.K., B.K., M.Z.; writing—review and editing, project administration, funding acquisition, M.K.

**Funding:** This research was funded by Ministry of Science and Higher Education, grant No NN 523 4942 34. Publication was funded by statutory activity of the Faculty of Building Services Hydro and Environmental Engineering, Warsaw University of Technology and of the Faculty of Environmental Engineering, Lublin University of Technology.

**Conflicts of Interest:** The authors declare no conflict of interest.

## References

1. Nawrocki, J.; Świetlik, J. Analysis of corrosion phenomena in water-pipe networks. *Ochrona Środowiska* **2011**, *33*, 27–40.
2. Rożej, A.; Cydzik-Kwiatkowska, A.; Kowalska, B.; Kowalski, D. Structure and microbial diversity of biofilms on different pipe materials of a model drinking water distribution systems. *World J. Microbiol. Biotechnol.* **2015**, *31*, 37–47. [[CrossRef](#)] [[PubMed](#)]

3. Kwietniewski, M.; Tłoczek, M.; Wysocki, L. (Eds.) *Zasady doboru rozwiązań materiałowo-konstrukcyjnych do budowy przewodów wodociagowych*; Izba Gospodarcza “Wodociągi Polskie”: Bydgoszcz, Poland, 2011.
4. Technical Standard PN-EN 545:2010. *Ductile Iron Pipes, Fittings, Accessories and Their Joints for Water Pipelines. Requirements and Test Methods*; Polish Committee for Standardization: Warsaw, Poland, 2010.
5. Musz, A.; Kowalska, B.; Widomski, M.K. Some issues concerning the problems of water quality modeling in distribution systems. *Ecol. Chem. Eng. S-Chemia I Inżynieria Ekologiczna S* **2009**, *16*, 175–184.
6. Neville, A. Effect of cement paste on drinking water. *Mater. Struct.* **2001**, *34*, 367–372. [[CrossRef](#)]
7. Świdarska-Bróz, M.; Wolska, M. Korozyjność wody wodociagowej a zjawiska zachodzące w systemie dystrybucji. *Gaz Woda Technika Sanitarna* **2003**, *1*, 10–15.
8. Świdarska-Bróz, M. Contributory factors in the potential of biofilm formation and growth in water distribution systems. *Ochrona Środowiska* **2010**, *32*, 7–13.
9. Douglas, B.D.; MERRIL, D.T. *Control of Water Quality Deterioration Caused by Corrosion of Cement-Mortar Pipe Linings*; AWWAFR: Denver, CO, USA, 1991.
10. Vik, E.A.; Hedberg, T. Corrosion and Corrosion Control in Drinking Water Systems. In Proceedings of the Corrosion Workshop and Seminar, Oslo, Norway, 19–21 March 1990.
11. Kowalska, B.; Kowalski, D.; Kwietniewski, M.; Miszta-Kruk, K.; Chudzicki, J. Heavy metal content of water exposed to cement lining in the water pipe. *Ochrona Środowiska* **2011**, *33*, 41–45.
12. Achternbosch, M.; Bräutigam, K.-R.; Hartlieb, N.; Kupsch, C.; Richers, U.; Stemmermann, P. *Heavy Metals in Cement and Concrete Resulting from the Co-Incineration of Wastes in Cement Kilns with Regard to the Legitimacy of Waste Utilisation*; Forschungszentrum Karlsruhe: Karlsruhe, Germany, 2003.
13. Van der Sloot, H.A. Comparison of the characteristic leaching behaviour of cements using standard (EN 196-1) cement mortar and an assessment on their long-term environmental behaviour in construction products during service life and recycling. *Cem. Concr. Res.* **2000**, *30*, 1079–1096. [[CrossRef](#)]
14. Guo, Q.; Toomuluri, J.P.; Eckert, J.O. Leachability of regulated metals from cement-mortar linings. *J. AWWA* **1998**, *90*, 62–73. [[CrossRef](#)]
15. Moudilou, E.; Bellotto, M.; Defosse, C.; Serclerat, I.; Baillif, P.; Touray, J.C. A dynamic leaching method for the assessment of trace metals released from hydraulic binders. *Waste Manag.* **2002**, *22*, 153–157. [[CrossRef](#)]
16. Berend, H.; Trouwborst, T. Cement- mortar pipes as a source of aluminium. *J. AWWA* **1999**, *91*, 91–100. [[CrossRef](#)]
17. WHO. *Guidelines for Drinking—Water Quality*, 3rd ed.; WHO: Geneva, Switzerland, 2008; Volume 1.
18. Regulation of the Polish Minister of Health. *April 20th 2010—About the Quality of Water Intended for Human Consumption (Dz.U. 72/2010, pos.466)*; Regulation of the Polish Minister of Health: Warsaw, Poland, 2010.
19. Le Corre, N. *Analysis of the Major Elements in Cement by ICP*; Jobin Yvon Entertainment: Palaiseau, France, 2010.
20. American Water Works Association. *Permeation and Leaching*; Paper issued on 15 August 2002; American Water Works Association: Denver, CO, USA, 2002.
21. Technical Standard PN-EN 12502-2. *Protection of Metallic Materials against Corrosion. Guidance on the Assessment of Corrosion Likelihood in Water Distribution and Storage Systems. Influencing Factors for Copper and Copper Alloys*; Polish Committee for Standardization: Warsaw, Poland, 2006.
22. Technical Standard PN-EN 12502-3. *Protection of Metallic Materials against Corrosion. Guidance on the Assessment of Corrosion Likelihood in Water Distribution and Storage Systems. Influencing Factors for Hot Dip Galvanised Ferrous Materials*; Polish Committee for Standardization: Warsaw, Poland, 2006.
23. Technical Standard PN-EN 12502-4. *Protection of Metallic Materials against Corrosion. Guidance on the Assessment of Corrosion Likelihood in Water Distribution and Storage Systems. Influencing Factors for Stainless Steels*; Polish Committee for Standardization: Warsaw, Poland, 2006.
24. Technical Standard PN-EN 12502-5. *Protection of Metallic Materials against Corrosion. Guidance on the Assessment of Corrosion Likelihood in Water Distribution and Storage Systems. Influencing Factors for Cast Iron, Unalloyed and Low Alloyed Steels*; Polish Committee for Standardization: Warsaw, Poland, 2006.
25. Kowalska, B.; Kowalski, D.; Kwietniewski, M.; Musz, A.; Wąsowski, J. Ocena wpływu wykładziny cementowej w rurociągach żeliwnych na jakość przesyłanej wody—badania wstępne. *Gaz Woda i Technika Sanitarna* **2010**, *4*, 13–17.

26. Genazzini, C.; Giaccio, G.; Ronco, A.; Zerbino, R. Cement-based materials as containment systems for ash from hospital waste incineration. *Waste Manag.* **2005**, *25*, 649–654. [[CrossRef](#)] [[PubMed](#)]
27. Giergieczny, Z.; Król, A. Immobilization of heavy metals (Pb, Cu, Cr, Cd, Zn, Mn) in the mineral additions containing concrete composites. *J. Hazard. Mater.* **2008**, *160*, 255–274. [[CrossRef](#)] [[PubMed](#)]



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