

## Article

# Determination of Chemical Species Dominating the Corrosivity of Japanese Tap Water by Multiple Regression Analysis

Yuji Nakamura <sup>1,\*</sup>, Yasuki Matsukawa <sup>1</sup>, Shinji Okazaki <sup>2</sup>  and Shukuji Asakura <sup>3</sup><sup>1</sup> SHINRYO CORPORATION, 41, Wadai, Tsukuba 300-4247, Japan; matsukawa.ya@shinryo.com<sup>2</sup> Graduate School of Engineering, Yokohama National University, 79-5, Tokiwadai, Hodogaya-ku, Yokohama 240-8501, Japan; okazaki-shinji-yp@ynu.ac.jp<sup>3</sup> Venture Academia Co., Ltd., 1-1-40, Suehiro, Tsurumi, Yokohama 230-0045, Japan; asakura@v-academia.jp

\* Correspondence: nakamura.yj@shinryo.com

**Abstract:** Japanese tap water is Ca<sup>2+</sup>-poor and SiO<sub>2</sub>-rich in comparison with that of other countries. Thus, there have been few studies on its corrosivity. We sampled tap waters at 70 different sites and in different seasons in Japan, subjected the samples to chemical analysis and measured localized corrosion depth and the total corrosion loss of carbon steel placed in these waters. The average corrosion rate  $v_{avg}$  and maximum localized corrosion rate  $v_{max}$  were calculated. The ratio of  $v_{max}$  to  $v_{avg}$ , which was defined as localized corrosion factor  $LCF$  ( $=v_{max}/v_{avg}$ ), was also studied. The multiple regression method was applied to obtain the dependence of  $v_{avg}$  (objective variable) on concentrations of chemical species (explanatory variables). In the same manner, the relation of  $v_{max}$  and  $LCF$  to concentrations of chemical species was derived. As a result, we showed that SiO<sub>2</sub> and SO<sub>4</sub><sup>2-</sup> mainly dominate the corrosivity of Japanese tap water. In particular, as SO<sub>4</sub><sup>2-</sup> increased,  $v_{avg}$  became larger and  $v_{max}$  became smaller. Also, as SiO<sub>2</sub> increased,  $v_{max}$  became larger and  $v_{avg}$  became smaller. The behavior of  $LCF$  was similar to that of  $v_{max}$ . The findings of this study will be useful for estimating the corrosivity of tap waters that have low Ca<sup>2+</sup> and high SiO<sub>2</sub> concentrations.

**Keywords:** carbon steel; tap water; water quality; corrosion; multiple regression analysis

**Citation:** Nakamura, Y.; Matsukawa, Y.; Okazaki, S.; Asakura, S. Determination of Chemical Species Dominating the Corrosivity of Japanese Tap Water by Multiple Regression Analysis. *Water* **2023**, *15*, 4299. <https://doi.org/10.3390/w15244299>

Academic Editor: Gang Wen

Received: 22 November 2023

Revised: 8 December 2023

Accepted: 11 December 2023

Published: 18 December 2023



**Copyright:** © 2023 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (<https://creativecommons.org/licenses/by/4.0/>).

## 1. Introduction

There are many steep mountains and volcanoes in Japan. Volcanoes are the cause of the high concentration of SiO<sub>2</sub> in river water. Meanwhile, the steep mountains result in fast-flowing rivers, and since CaCO<sub>3</sub> dissolves slowly into water, the concentration of Ca<sup>2+</sup> is low, causing the tap water to be soft. Hori et al. [1] studied the hardness of tap waters in Japan and compared the results with those of European countries. They reported that total hardness at 16 places in France was 178 ± 68 mg/L as CaCO<sub>3</sub>, at 11 places in Germany was 187 ± 71 mg/L as CaCO<sub>3</sub> and at 665 places in Japan was 48.9 ± 25.8 mg/L as CaCO<sub>3</sub>. A more detailed study of tap water in European countries was conducted by Banks et al. [2,3], who showed that the average calcium hardness of 579 tap waters sampled in European countries was 60.1 mg/L (=149.9 mg/L as CaCO<sub>3</sub>). Almost no report of calcium hardness in the US could be found; however, there have been some studies that investigated the hardness of river water in the US. Brigges et al. found that the mean hardness of river water at 344 sites was 223 mg/L as CaCO<sub>3</sub> [4]. In general, tap water is produced by treating river water, so it is reasonable to assume that tap water inherits the properties of river water, which explains why tap water in the US has high calcium hardness. On the contrary, Japanese tap water has low calcium hardness compared to that in European countries and America [1–5].

The relationship between corrosion rates and components in tap water has often been studied in laboratory settings. It is considered that Cl<sup>-</sup> and SO<sub>4</sub><sup>2-</sup> are corrosive and that HCO<sub>3</sub><sup>-</sup> inhibits corrosion [6–13]. The effects of Cl<sup>-</sup> and SO<sub>4</sub><sup>2-</sup> in the concentration range of fresh water were reported by Takasaki et al. [10] and Agata et al. [12]. They found that

when the concentrations of  $\text{Cl}^-$  and  $\text{SO}_4^{2-}$  are high enough, the corrosion rate almost becomes constant, while the rate increases when their concentrations are low [10,11]. In another similar study, Fujii et al. [13] added sodium chloride and sodium sulfate to tap water and found that the corrosivity did not change when the sum of  $\text{Cl}^-$  and  $\text{SO}_4^{2-}$  concentrations was above 20 mg/L.

Conventionally, the corrosivity of tap water is evaluated by corrosion indexes such as the Langelier Index (LI) [14,15], Ryzner Stability Index (RSI) [16] and Larson–Skold Index (LSI) [6,14–18]. However, according to the authors’ experience, these indexes do not always reflect actual corrosivity, particularly for Japanese tap water [19]. Nakamura et al. show that there is not always much of a relationship between the LI and RSI of Japanese tap water, average corrosion and localized corrosion of carbon steel [19]. We assume that this is due to the fact that conventional corrosion indexes were obtained on the basis of corrosion tests performed with tap water of high calcium hardness.

Recently, corrosion studies have frequently used multivariate analysis for multiple regression [20,21]. Sobue et al. [20] applied multiple regression analysis to the relationship between chemical components and electrochemical impedance to determine the factors that significantly affect copper corrosion in tap water and found that free carbon dioxide ( $\text{CO}_2 \text{ aq}$ ) was the most accelerating corrosion factor. So et al. [21] used regression analysis to study corrosion factors in district heating water and presented a regression formula for predicting the corrosion depth as a function of pH, concentration of dissolved oxygen and time. Their study showed that multiple regression analysis can be applied to corrosion studies to determine the components in aqueous solutions that dominate corrosion [21].

In the present study, water quality measurements and corrosion tests for carbon steel were performed for 70 different tap waters in Japan sampled at different locations and in different months. The sources of the sampled tap water were ground water, surface water or their mixture. Moreover, multiple regression analysis for clarifying the relationship between chemical species and corrosion rates was conducted to determine the chemical species in tap water that dominate corrosivity.

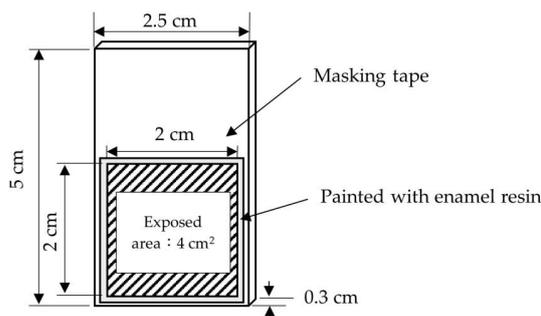
**2. Materials and Methods**

*2.1. Materials*

Carbon steel ASTM A283 (JFE Steel Corporation, Tokyo, Japan) 50 mm × 25 mm × 3 mm polished with P400 [22] was used for the test specimens. The chemical composition of the specimens is shown in Table 1. The specimens were degreased with ethanol and acetone and covered with chemical-resistant adhesive tape except for the exposed areas. The boundary between the metal and the adhesive tape was painted with enamel resin. The exposed metal area was 4 cm<sup>2</sup> (2 cm × 2 cm). A schematic drawing of a test specimen is illustrated in Figure 1.

**Table 1.** Chemical composition of test piece (carbon steel, SS400).

Element	Fe	C	Si	Mn	P	S
Content [wt%]	99.3	0.08	0.03	0.37	0.08	0.14



**Figure 1.** Schematic of test piece.

## 2.2. Sampling and Measurement of Tap Waters Used for Corrosion Tests

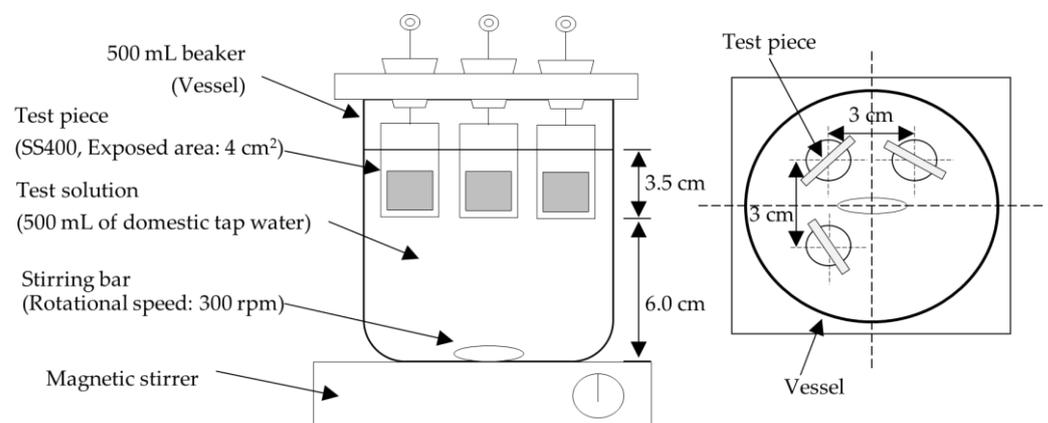
Seventy different Japanese tap waters were sampled and their pH, electrical conductivity,  $\text{HCO}_3^-$ ,  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{Cl}^-$ ,  $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$  and  $\text{SiO}_2$  were measured. The pH was measured with a glass electrode. The electrical conductivity was measured by the method of two platinum electrodes whose surfaces were covered with platinum black. The concentration of  $\text{HCO}_3^-$  was determined from the value of M-alkalinity, which was determined by titration of bromocresol green–methyl red ethanol solution and sulfuric acid. The concentrations of  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{Cl}^-$ ,  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$  were quantitatively determined by ion chromatography. The concentration of  $\text{SiO}_2$  was quantitatively analyzed by molybdenum yellow absorptiometry. Since Japanese tap water is treated with hypochlorous acid (0.1–1 mg/L) [23], the number of bacteria was not measured. Therefore, microbial corrosion was not taken into account. The corrosivity of these tap waters for carbon steel was evaluated by corrosion tests as follows.

## 2.3. Method of Immersion Test

The corrosivity of the sampled tap waters was evaluated by immersion tests. The testing time in this study was set according to common standards. The corrosion testing guidelines (i.e., ASTM G31-72 [24], Standard Practice for Laboratory Immersion Corrosion Testing of Metals and NACE Standard TM-01-69 [25], Test Method—Laboratory Corrosion Testing of Metals for the Process Industries) recommend that test periods (time,  $t$  (hours)) are longer than the following Equation (1):

$$t = 50/v \quad (1)$$

where  $v$  is conventional corrosion rate ( $\text{mm y}^{-1}$ ) in test solutions. It is known that the corrosion rate of carbon steel immersed in neutral solutions (pH between 4 and 10) is about  $0.1 \text{ mm y}^{-1}$  [26]. Therefore, at least 500 h is required for corrosion tests. For this reason, the specimens were immersed in 500 mL of the test solution for approximately 720 h. The experimental apparatus is shown in Figure 2. The temperature of the test solution was set at  $25 \text{ }^\circ\text{C}$ . The solution was stirred at 300 rpm using a stirring bar controlled with a magnetic stirrer in the open-to-air state. Three specimens were placed in a vessel. The ratio of solution volume to specimen area was approximately  $40 \text{ mL/cm}^2$ . The corrosion test guidelines mentioned above require a solution volume to specimen area greater than  $20 \text{ mL/cm}^2$ . The ratio in this study meets this criterion. At the time of sampling and during the corrosion test, dissolved oxygen was not measured, since it was considered that the oxygen in the solution was in the saturated state by air because of its continuous stirring.



**Figure 2.** Schematic of the experimental apparatus.

#### 2.4. Measurement of Corrosion

The corrosion products that formed on a test piece were removed with 7% hydrochloric acid containing 0.5% corrosion inhibitor for acid cleaning (Asahi Chemical Co., Ltd., Osaka, Japan, IBIT<sup>®</sup> No. 2AS, Aqueous solution containing quaternary ammonium salts, nonionic surfactants, isopropyl alcohol, and benzyl trimethylammonium chloride). The average weight loss of the three specimens divided by the testing time was defined as  $v_{avg}$ . The localized corrosion depths were measured with a depth gauge (Mitsutoyo, Kanagawa, Japan, point micrometer). The depths of the five most significantly corroded areas were measured. The maximum value among them divided by the testing time was defined as  $v_{max}$ . In addition, the ratio of  $v_{avg}$  to  $v_{max}$  was calculated and defined as the localized corrosion factor (LCF). The above procedures are shown schematically in Figure 3.



**Figure 3.** Process from surface treatment to measurement of corrosion.

#### 2.5. Multiple Regression Analysis

The corrosive effect of the chemical species was evaluated on the basis of the actual corrosion rates and qualities of tested waters by multiple regression analysis [27]. The regression equation obtained from multiple regression analysis is given by

$$y = \alpha + \beta_1x_1 + \beta_2x_2 + \dots + \beta_px_p + \varepsilon \quad (2)$$

where  $y$  is the objective variable, corrosivity;  $p$  is the number of independent variables;  $\alpha$  is the regression constant;  $\beta$  is the regression coefficient for  $x_p$ ;  $x_p$  is the explanatory variable, the concentrations of various chemical species; and  $\varepsilon$  is random error [27].

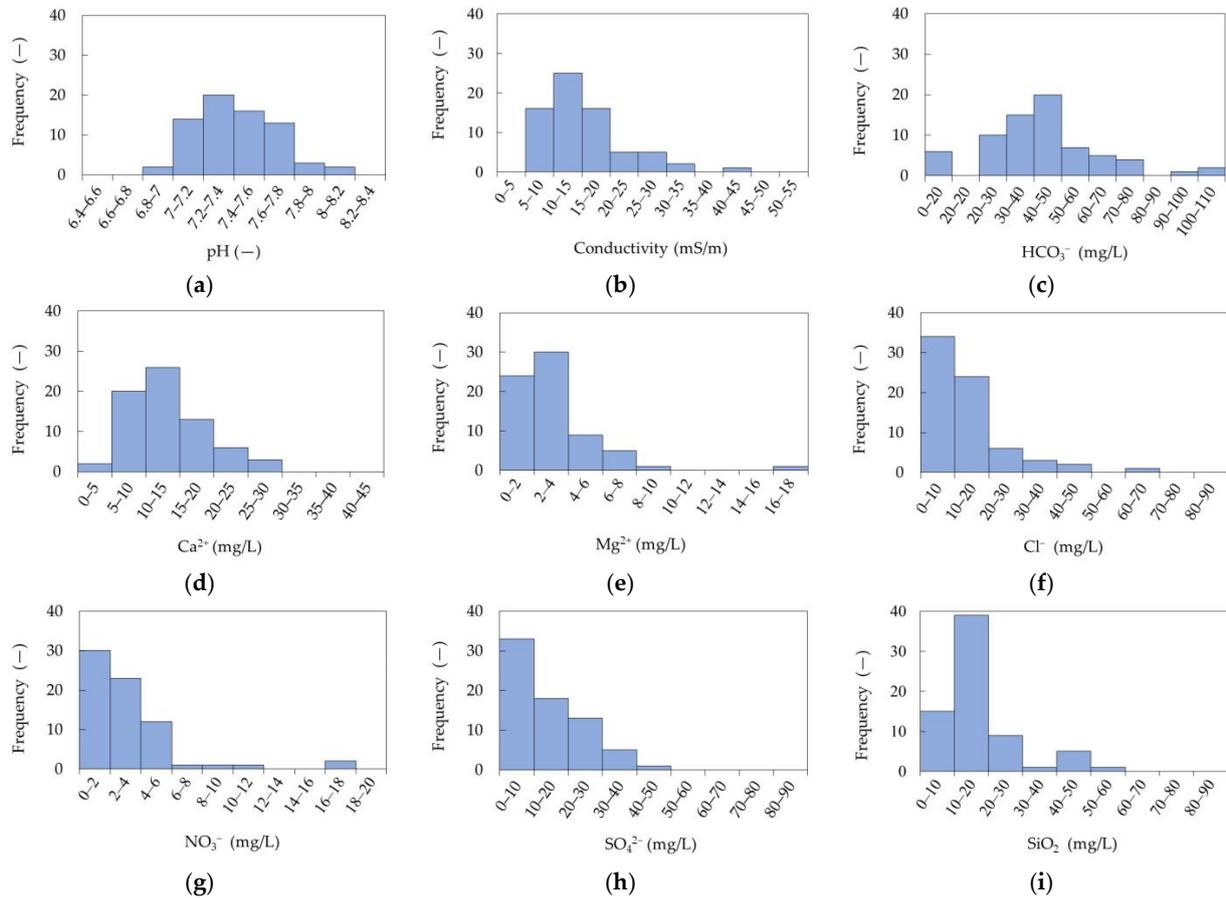
In this study, several models with different numbers of explanatory variables were tested and the coefficient of determination ( $R^2$ ) with standard error (SE) of each model was compared. Based on the value of the regression variable  $x$ , the impact of that component on the corrosion rate was evaluated. Units of water constituents are given in mol/L and are normalized to a maximum value of 1. This means that the higher the regression coefficient  $x$ , the larger the impact on corrosion. The model for the regression analysis was tested for eight water species,  $H^+$ ,  $Ca^{2+}$ ,  $Mg^{2+}$ ,  $HCO_3^-$ ,  $Cl^-$ ,  $SO_4^{2-}$ ,  $NO_2^-$  and  $SiO_2$  for the regression constant  $\beta$ , with the value of  $\beta$  reduced in steps until all variance expansion factors (VIFs) were less than 2 [28]. VIF calculations and multiple regression analysis were performed using Microsoft Excel (ver.2311, Microsoft Corporation, Redmond, WA, USA).

### 3. Results

#### 3.1. Quality of Japanese Tap Waters

Histograms of the quality of Japanese tap waters and those of average value, maximum value and minimum value are shown in Figure 4 and Table 2. The concentrations of components in Japanese tap water are widely distributed. Focusing on the concentrations of  $Ca^{2+}$  and  $SiO_2$ , the tap waters collected in this study had  $Ca^{2+}$  values in the range of 3.6–28.4 mg/L and  $SiO_2$  in the range of 5–59 mg/L. In addition, the conductivity was distributed in the range of 5.7–43.9 mS/m. These results reflect the fact that there are many steep mountains and volcanoes in Japan, which typically lead to short rivers with water rich in  $SiO_2$  but low in calcium compounds due to the high flow rate and low dissolution rate of  $CaCO_3$  into water. As a result, Japanese tap waters are soft with lower levels of dissolved minerals than those in other countries. The solutions used in these corrosion tests

reflect the characteristics of Japanese tap water [1]. Figure 5 shows the correlations among chemical species in tap water. In multiple regression analysis, it is desirable to reduce the correlations between the explanatory variables. Therefore, in the analysis, a few models with smaller VIFs were created by referring to Figure 5, which represents the strength of the correlations.



**Figure 4.** Histogram of concentrations of chemical compositions in 70 different Japanese tap waters. (a) pH, (b) conductivity, (c) HCO<sub>3</sub><sup>-</sup>, (d) Ca<sup>2+</sup>, (e) Mg<sup>2+</sup>, (f) Cl<sup>-</sup>, (g) SO<sub>4</sub><sup>2-</sup>, (h) NO<sub>3</sub><sup>-</sup>, (i) SiO<sub>2</sub>.

**Table 2.** Water quality of 70 different tap waters in Japan.

Species	Unit	Average	Maximum	Minimum
pH (25 °C)	-	7.46	8.1	7.0
Conductivity	mS/m	15.3	43.9	5.7
HCO <sub>3</sub> <sup>-</sup>	mg/L	43.6	106.1	13.4
	mmol/L	0.714	1.740	0.220
Ca <sup>2+</sup>	mg/L	13.0	28.4	3.6
	mmol/L	0.324	0.709	0.090
Mg <sup>2+</sup>	mg/L	3.1	17.2	0.7
	mmol/L	0.129	0.709	0.030
Cl <sup>-</sup>	mg/L	13.9	62	2
	mmol/L	0.391	1.749	0.056
SO <sub>4</sub> <sup>2-</sup>	mg/L	14.9	41.4	1.3
	mmol/L	0.155	0.431	0.014
NO <sub>3</sub> <sup>-</sup>	mg/L	3.1	18	0.1
	mmol/L	0.050	0.290	0.002
SiO <sub>2</sub>	mg/L	17.1	59	2
	mmol/L	0.284	0.982	0.033

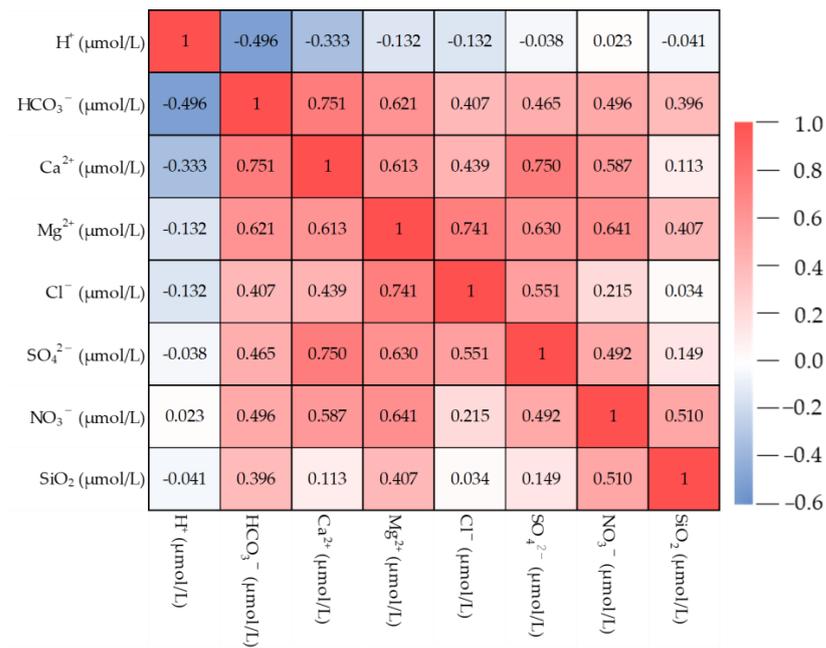


Figure 5. Correlations between water quality components in 70 different tap waters.

### 3.2. Corrosivity of Japanese Tap Waters

Carbon steel specimens corroded in all 70 Japanese tap waters. The corrosion situation of some of the specimens was as shown in Figure 6. Corrosion was observed under the corrosion products, and the corrosion situation was different. Case 1 and Case 2 corroded almost uniformly; in contrast, uniform corrosion and localized corrosion pits were observed in Case 3. This result means that the corrosion situation of carbon steel depends on the water quality. The average corrosion rate  $v_{avg}$ , localized corrosion rate  $v_{max}$  and localized corrosion factor LCF obtained from corrosion tests are shown in Figure 7. The highest value of  $v_{avg}$  is  $0.541 \text{ mm y}^{-1}$ , the lowest is  $0.105 \text{ mm y}^{-1}$ , the mean is  $0.302 \text{ mm y}^{-1}$  and the mode is 0.28 to  $0.32 \text{ mm y}^{-1}$ . The highest value of  $v_{max}$  is  $4.16 \text{ mm y}^{-1}$ , the lowest is  $0.58 \text{ mm y}^{-1}$ , the mean is  $1.92 \text{ mm y}^{-1}$  and the mode is 0.5 to  $1.0 \text{ mm y}^{-1}$ . The highest value of LCF is 20.6, the lowest is 1.68, the mean is 7.08 and the mode is 2 to 4. Most notable among these results is the presence of tap water, which causes localized corrosion that progresses at a rate 20.6 times faster than average corrosion. These results show that corrosion rates are sensitive to the effects of water quality. Focusing on the shape of the distribution,  $v_{avg}$  was normally distributed. In contrast, the shape of  $v_{max}$  and LCF were skew distributed. The results indicate that most tap waters in Japan have low localized corrosivity, although a few have significantly high localized corrosivity.

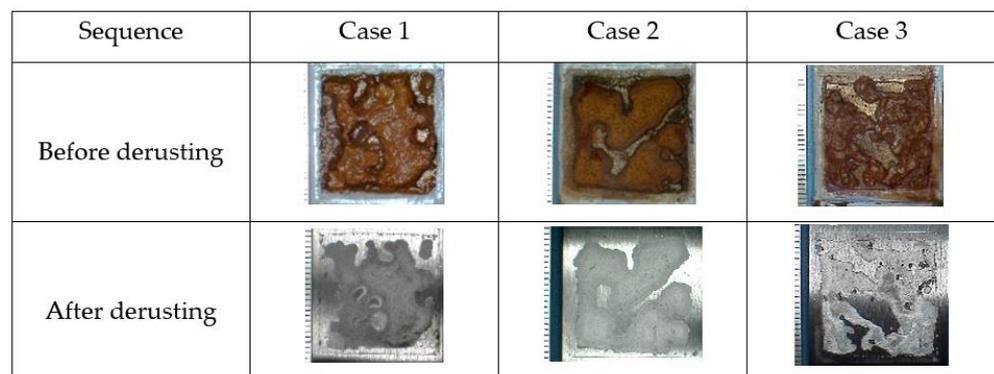
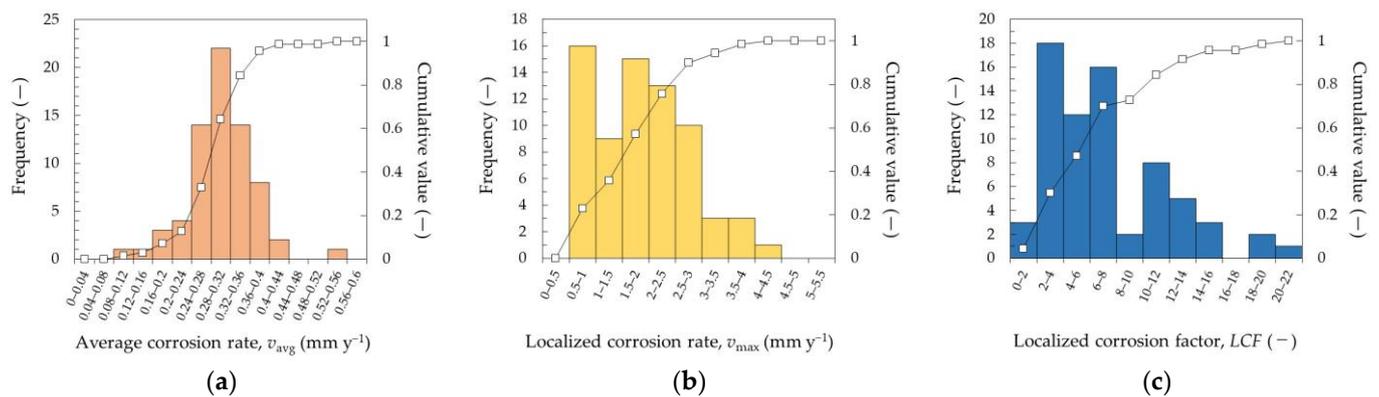


Figure 6. Three examples of the appearance of a carbon steel specimen immersed in tap water for 720 h.



**Figure 7.** Histogram of corrosion rates of carbon steel in 70 different tap waters in Japan. (a) Average corrosion rate,  $v_{avg}$ . (b) Localized corrosion rate,  $v_{max}$ . (c) Localized corrosion factor, LCF.

### 3.3. Relationships between Water Quality and Corrosivity

The correlation coefficients (R) between  $v_{avg}$ ,  $v_{max}$  and LCF for each chemical species in the tested waters are shown in Table 3. A close examination of these results clarifies the following. First, regarding  $v_{avg}$ ,  $H^+$ ,  $Ca^{2+}$ ,  $Cl^-$  and  $SO_4^{2-}$  promoted corrosion because the correlation coefficients were positive. Meanwhile,  $HCO_3^-$ ,  $Mg^{2+}$ ,  $NO_3^-$  and  $SiO_2$  inhibited corrosion because the correlation coefficients were negative. Next, with regard to  $v_{max}$ , since the correlation coefficients were positive,  $HCO_3^-$  and  $SiO_2$  promoted corrosion.  $Ca^{2+}$ ,  $Mg^{2+}$ ,  $Cl^-$ ,  $SO_4^{2-}$  and  $NO_3^-$  inhibited corrosion because the correlation coefficients were negative. Finally, regarding LCF, since the correlation coefficients were positive,  $HCO_3^-$ ,  $NO_3^-$  and  $SiO_2$  promoted localized corrosion.  $H^+$ ,  $Ca^{2+}$ ,  $Mg^{2+}$ ,  $Cl^-$  and  $SO_4^{2-}$  inhibited localized corrosion because the correlation coefficients were negative.

**Table 3.** Correlation coefficient (R) of corrosivity with chemical components in 70 different tap waters in Japan.

Corrosivity	$H^+$	$HCO_3^-$	$Ca^{2+}$	$Mg^{2+}$	$Cl^-$	$SO_4^{2-}$	$NO_3^-$	$SiO_2$
$v_{avg}$	0.124	−0.166	0.102	−0.031	0.226	0.302	−0.243	−0.468
$v_{max}$	−0.081	0.005	−0.342	−0.211	−0.328	−0.432	−0.152	0.401
LCF	−0.145	0.066	−0.289	−0.111	−0.316	−0.440	0.030	0.506

Focusing on the absolute values of the correlation coefficients, the values for  $SO_4^{2-}$  and  $SiO_2$  were larger than those for the other species. This result suggests that these are dominant for the corrosion of carbon steel in tap water. The role of  $SiO_2$  was also taken into consideration. Previous papers have reported that  $SiO_2$  formed a film on zinc and copper [29]. If a similar film was formed on the iron surface, the corrosion inhibitive nature of  $SiO_2$  could be explained. The suppressive effect of the  $SiO_2$  obtained in the authors' experiment was proved to be consistent with this assumption. Yuasa et al. [30] reported that the corrosion of carbon steel in artificial fresh water was inhibited by  $SiO_2$ . On the other hand, attention should be paid to the fact that  $v_{max}$  was an increasing function with respect to  $SiO_2$ . That is,  $SiO_2$  promotes localized corrosion. Conventionally, it has been believed that  $HCO_3^-$  inhibits corrosion [6,7,10,11], but in fact it did not affect corrosion.

### 3.4. Extraction of Dominant Species Affecting Corrosivity of Waters

#### 3.4.1. Constructing Multiple Regression Models

The explanatory variables were deleted step by step by considering the highest VIF. This operation was repeated with all VIFs that were less than two. First, model A consisted of  $H^+$ ,  $HCO_3^-$ ,  $Ca^{2+}$ ,  $Mg^{2+}$ ,  $Cl^-$ ,  $SO_4^{2-}$ ,  $NO_3^-$  and  $SiO_2$ .  $Ca^{2+}$  showed the highest VIF of 6.18. Therefore,  $Ca^{2+}$  was excluded from model A. This process brought model B. In the same manner, model C and model D were obtained. Finally, in model D, the VIF was

less than two. Table 4 shows the results of this process. Multiple regression analysis was performed on models A to D with  $v_{avg}$ ,  $v_{max}$  and LCF as the objective variables.

**Table 4.** VIF values among water species in each multiple regression model.

Model	H <sup>+</sup>	HCO <sub>3</sub> <sup>-</sup>	Ca <sup>2+</sup>	Mg <sup>2+</sup>	Cl <sup>-</sup>	SO <sub>4</sub> <sup>2-</sup>	NO <sub>3</sub> <sup>-</sup>	SiO <sub>2</sub>
A	1.63	4.30	6.73	5.81	3.53	3.40	3.22	2.22
B	1.57	2.64	–	5.81	3.48	1.94	2.67	1.62
C	1.57	2.53	–	–	1.55	1.92	1.98	1.49
D	1.03	–	–	–	1.47	1.84	1.79	1.39

### 3.4.2. Multiple Regression Analysis on Chemical Species and $v_{avg}$

The R<sup>2</sup> and the regression coefficient for  $v_{avg}$  are shown in Table 5. The R<sup>2</sup> varied from 0.427 to 0.432. The standard error (SE) was between 0.096 and 0.098. The explanatory variables with large regression coefficients were SiO<sub>2</sub>, NO<sub>3</sub><sup>-</sup> and SO<sub>4</sub><sup>2-</sup>. In addition, the *p* values obtained from the multiple regression analysis are shown in Table 6. The *p* value means whether the coefficients of the explanatory variables are statistically significant or not. In this study, a *p* value of 0.05 or less was considered significant. Considering the *p* value of models C and D, it seems that SiO<sub>2</sub>, NO<sub>3</sub><sup>-</sup> and SO<sub>4</sub><sup>2-</sup> affect  $v_{avg}$  significantly. This result shows that  $v_{avg}$  mainly dominated with these chemical species. Additionally, a negligible effect of Cl<sup>-</sup> and HCO<sub>3</sub><sup>-</sup> on  $v_{avg}$  was found. The signs of SiO<sub>2</sub> and NO<sub>3</sub><sup>-</sup> were negative and that of SO<sub>4</sub><sup>2-</sup> was positive. This means that SiO<sub>2</sub> and NO<sub>3</sub><sup>-</sup> decrease the average corrosion rate, while SO<sub>4</sub><sup>2-</sup> increases it.

**Table 5.** R<sup>2</sup> and regression coefficient in the multiple regression models for  $v_{avg}$ .

Model	R <sup>2</sup>	SiO <sub>2</sub>	NO <sub>3</sub> <sup>-</sup>	SO <sub>4</sub> <sup>2-</sup>	Cl <sup>-</sup>	Mg <sup>2+</sup>	Ca <sup>2+</sup>	HCO <sub>3</sub> <sup>-</sup>	H <sup>+</sup>	b	SE
A	0.432	−0.224	−0.189	0.254	0.061	−0.018	0.020	−0.079	0.051	0.555	0.098
B	0.432	−0.230	−0.182	0.262	0.060	−0.018	–	−0.069	0.049	0.559	0.097
C	0.432	−0.232	−0.187	0.261	0.052	–	–	−0.071	0.049	0.560	0.096
D	0.427	−0.247	−0.208	0.251	0.039	–	–	–	0.076	0.535	0.096

**Table 6.** *p* values in each multiple regression model for  $v_{avg}$ .

Model	SiO <sub>2</sub>	NO <sub>3</sub> <sup>-</sup>	SO <sub>4</sub> <sup>2-</sup>	Cl <sup>-</sup>	Mg <sup>2+</sup>	Ca <sup>2+</sup>	HCO <sub>3</sub> <sup>-</sup>	H <sup>+</sup>
A	1.85 × 10 <sup>-2</sup>	0.123	8.31 × 10 <sup>-3</sup>	0.614	0.929	0.899	0.548	0.445
B	4.66 × 10 <sup>-3</sup>	0.099	3.91 × 10 <sup>-4</sup>	0.619	0.929	–	0.502	0.448
C	2.78 × 10 <sup>-3</sup>	0.048	3.39 × 10 <sup>-4</sup>	0.515	–	–	0.478	0.445
D	1.03 × 10 <sup>-3</sup>	0.021	3.98 × 10 <sup>-4</sup>	0.614	–	–	–	0.145

### 3.4.3. Multiple Regression Analysis on Chemical Species and $v_{max}$

The R<sup>2</sup> and the regression coefficients for  $v_{avg}$  are shown in Table 7. The R<sup>2</sup> varied from 0.474 to 0.461. The SE was between 0.163 and 0.165. Roughly, the explanatory variables with large regression coefficients were SiO<sub>2</sub>, NO<sub>3</sub><sup>-</sup> and SO<sub>4</sub><sup>2-</sup>. In addition, the *p* values are shown in Table 8. In models B, C and D, *p* values less than 0.05 were obtained for SiO<sub>2</sub>, SO<sub>4</sub><sup>2-</sup> and NO<sub>3</sub><sup>-</sup>. As with  $v_{avg}$ ,  $v_{max}$  mainly dominated with these chemical species. Focusing on the sign of the regression coefficient, SiO<sub>2</sub> was positive, while SO<sub>4</sub><sup>2-</sup> and NO<sub>3</sub><sup>-</sup> were negative. Therefore, it can be said that SiO<sub>2</sub> increases the localized corrosion rate, while SO<sub>4</sub><sup>2-</sup> and NO<sub>3</sub><sup>-</sup> decrease it.

**Table 7.**  $R^2$  and regression coefficient in the multiple regression models for  $v_{max}$ .

Model	$R^2$	SiO <sub>2</sub>	NO <sub>3</sub> <sup>-</sup>	SO <sub>4</sub> <sup>2-</sup>	Cl <sup>-</sup>	Mg <sup>2+</sup>	Ca <sup>2+</sup>	HCO <sub>3</sub> <sup>-</sup>	H <sup>+</sup>	b	SE
A	0.474	0.592	-0.329	-0.283	-0.172	-0.038	-0.115	0.255	-0.012	0.446	0.165
B	0.472	0.627	-0.365	-0.328	-0.162	-0.037	-	0.195	-0.002	0.424	0.164
C	0.472	-0.232	-0.187	0.261	0.052	-	-	-0.071	0.049	0.560	0.163
D	0.461	0.661	-0.320	-0.301	-0.143	-	-	-	-0.075	0.496	0.163

**Table 8.**  $p$  values in each multiple regression model for  $v_{max}$ .

Model	SiO <sub>2</sub>	NO <sub>3</sub> <sup>-</sup>	SO <sub>4</sub> <sup>2-</sup>	Cl <sup>-</sup>	Mg <sup>2+</sup>	Ca <sup>2+</sup>	HCO <sub>3</sub> <sup>-</sup>	H <sup>+</sup>
A	$3.69 \times 10^{-4}$	0.113	$7.79 \times 10^{-2}$	0.405	0.913	0.665	0.254	0.915
B	$1.43 \times 10^{-5}$	$5.23 \times 10^{-2}$	$7.40 \times 10^{-3}$	0.427	0.913	-	0.263	0.984
C	$6.52 \times 10^{-6}$	$2.03 \times 10^{-2}$	$6.48 \times 10^{-3}$	0.189	-	-	0.259	0.982
D	$1.01 \times 10^{-6}$	$3.72 \times 10^{-2}$	$1.07 \times 10^{-2}$	-	-	-	0.277	0.395

### 3.4.4. Multiple Regression Analysis on Chemical Species and LCF

The  $R^2$  and the regression coefficients for LCF are shown in Table 9, and the  $p$  values are shown in Table 10. The results show similar trends to  $v_{max}$ . The SE was between 0.152 and 0.155. The  $R^2$  was between 0.554 and 0.557, which is higher than that of  $v_{avg}$  and  $v_{max}$ . The explanatory variables with large regression coefficients were SiO<sub>2</sub> and SO<sub>4</sub><sup>2-</sup>. LCF mainly dominated with these chemical species. Moreover, being focused on  $p$  values, SiO<sub>2</sub> and SO<sub>4</sub><sup>2-</sup> values were below 0.05 and significantly smaller. This indicates that it may be possible to determine the corrosivity based on only two variables. Focusing on the sign of the regression coefficient, SiO<sub>2</sub> was positive and SO<sub>4</sub><sup>2-</sup> was negative. The larger the LCF, the more localized the corrosion, so it can be assumed that SiO<sub>2</sub> localizes the corrosion, while SO<sub>4</sub><sup>2-</sup> causes uniform corrosion.

**Table 9.**  $R^2$  and regression coefficient in the multiple regression models for LCF.

Model	$R^2$	SiO <sub>2</sub>	NO <sub>3</sub> <sup>-</sup>	SO <sub>4</sub> <sup>2-</sup>	Cl <sup>-</sup>	Mg <sup>2+</sup>	Ca <sup>2+</sup>	HCO <sub>3</sub> <sup>-</sup>	H <sup>+</sup>	b	SE
A	0.557	0.617	-0.003	-0.421	-0.145	0.030	-0.107	0.125	-0.129	0.394	0.155
B	0.556	0.651	-0.037	-0.463	-0.135	0.031	-	0.069	-0.119	0.373	0.154
C	0.556	0.654	-0.029	-0.462	-0.121	-	-	0.073	-0.119	0.371	0.153
D	0.554	0.669	-0.008	-0.452	-0.108	-	-	-	-0.147	0.398	0.152

**Table 10.**  $p$  values in each multiple regression model for LCF.

Model	SiO <sub>2</sub>	NO <sub>3</sub> <sup>-</sup>	SO <sub>4</sub> <sup>2-</sup>	Cl <sup>-</sup>	Mg <sup>2+</sup>	Ca <sup>2+</sup>	HCO <sub>3</sub> <sup>-</sup>	H <sup>+</sup>
A	$8.74 \times 10^{-5}$	0.987	$5.95 \times 10^{-3}$	0.454	0.925	0.666	0.548	0.228
B	$2.21 \times 10^{-6}$	0.830	$9.44 \times 10^{-5}$	0.479	0.923	-	0.669	0.249
C	$6.94 \times 10^{-7}$	0.846	$7.97 \times 10^{-5}$	0.337	-	-	0.646	0.246
D	$1.58 \times 10^{-7}$	0.956	$7.21 \times 10^{-5}$	-	-	-	0.376	0.077

## 4. Conclusions

Tap waters sampled at 70 different places and in different seasons in Japan were subjected to chemical analysis, and the corrosion of carbon steel in these waters was studied. The average corrosion rate  $v_{avg}$ , the maximum  $v_{max}$  and the LCF ( $=v_{max}/v_{avg}$ ) were discussed.  $v_{avg}$  was in the range of 0.105 mm y<sup>-1</sup> to 0.54 mm y<sup>-1</sup>,  $v_{max}$  was 0.58 mm y<sup>-1</sup> to 4.16 mm y<sup>-1</sup> and LCF was 1.68 to 20.1. Multiple regression analysis was carried out by setting chemical species as explanatory variables and LCF and corrosion rates as objective variables. We succeeded in showing that SiO<sub>2</sub> and SO<sub>4</sub><sup>2-</sup> dominate the corrosivity of Japanese tap water. In particular, as SO<sub>4</sub><sup>2-</sup> increased,  $v_{avg}$  became larger and  $v_{max}$  became smaller. Also, as SiO<sub>2</sub> increased,  $v_{max}$  became larger and  $v_{avg}$  became smaller. LCF behaved

similarly to  $v_{\max}$ . The authors expect that the findings of this study will be useful for estimating the corrosivity of tap waters with low  $\text{Ca}^{2+}$  and high  $\text{SiO}_2$  concentrations.

**Author Contributions:** Conceptualization, Y.N. and S.A.; methodology, Y.N.; validation, Y.N.; formal analysis, Y.N.; investigation, Y.N.; resources, Y.N.; data curation, Y.N.; writing—original draft preparation, Y.N.; writing—review and editing, S.O. and S.A.; visualization, Y.N.; supervision, S.O. and Y.M.; project administration, S.O. and Y.M.; funding acquisition, Y.M. All authors have read and agreed to the published version of the manuscript.

**Funding:** This research received no external funding.

**Data Availability Statement:** The data presented in this study are available on request from the corresponding author.

**Conflicts of Interest:** Authors Yuji Nakamura and Yasuki Matsukawa were employed by the company SHINRYO CORPORATION. Author Shukuji Asakura was employed by the company Venture Academia Co., Ltd. The remaining authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest. The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

## References

1. Hori, M.; Shozugawa, K.; Sugimori, K.; Watanabe, Y. A survey of monitoring tap water hardness in Japan and its distribution patterns. *Sci. Rep.* **2021**, *11*, 13546. [[CrossRef](#)] [[PubMed](#)]
2. Banks, D.; Birke, M.; Flem, B.; Reimann, C. Inorganic chemical quality of European tap-water: 1. Distribution of parameters and regulatory compliance. *Appl. Geochem.* **2015**, *59*, 200–210. [[CrossRef](#)]
3. Flem, B.; Reimann, C.; Birle, D.; Banks, D.; Filzmoser, P.; Frengstad, B. Inorganic chemical quality of European tap-water: 2. Geographical distribution. *Appl. Geochem.* **2015**, *59*, 211–224. [[CrossRef](#)]
4. Briggs, C.J.; Ficke, F.J. *Quality of Rivers of the United States, 1975 Water Year—Based on the National Stream Quality Accounting Network*; U.S. GEOLOGICAL SURVEY: Reston, VA, USA, 1977; pp. 24–25.
5. Ogura, K. The Water Quality in the world and Japan. *Heat. Air-Cond. Sanit. Eng.* **2013**, *3*, 17–19.
6. Larson, T.E.; Skold, R.V. Laboratory Studies Relating Mineral Quality of Water to Corrosion of Steel and Cast Iron. *Corrosion* **1958**, *14*, 285–288. [[CrossRef](#)]
7. Takasaki, S.; Yamada, Y. Effects of temperature and aggressive anions on corrosion of carbon steel in potable water. *Corros. Sci.* **2007**, *49*, 240–247. [[CrossRef](#)]
8. Haruna, T.; Domoto, K.; Shibata, T. Effect of Sulfate Ion on Corrosion of Carbon Steel in Carbonate/Bicarbonate Solutions. *Zairyo-to-Kankyo* **2002**, *51*, 350–355. [[CrossRef](#)]
9. Fukaya, Y.; Akashi, M. Passivation Behavior of Carbon Steel in Aqueous Carbonate Solutions. *Zairyo-to-Kankyo* **2002**, *56*, 521–528. [[CrossRef](#)]
10. Takasaki, S.; Ueki, H.; Yamamoto, D. Corrosion of mild steel in demineralized water. In Proceedings of the 76th Japan Corrosion Conference, Tokyo, Japan, 12 May 1976.
11. Agata, K.; Kazama, M.; Nakajima, H.; Hosoya, K.; Matsukawa, Y. Corrosion of building equipment and water quality. In Proceedings of the JSCE Materials and Environments 2001, Ibaraki, Japan, 30 May 2001.
12. Videm, K.; Koren, A.M. Corrosion, Passivity, and Pitting of Carbon Steel in Aqueous Solutions of  $\text{HCO}_3^-$ ,  $\text{CO}_2$ , and  $\text{Cl}^-$ . *Corrosion* **1993**, *49*, 746–755. [[CrossRef](#)]
13. Fujii, T.; Kodama, T.; Baba, H. Influence of water quality and flow conditions on corrosion of carbon steel pipes in fresh water. *Boshoku-Gijyutsu Present. Zair.-Kankyo* **1982**, *31*, 637–642.
14. Langelier, W.F. The analytical control of anticorrosion water treatment. *J. Am. Water Works Assn.* **1936**, *28*, 1500–1521. [[CrossRef](#)]
15. Langelier, W.F. Effect of Temperature on the pH of Natural Waters. *J. Am. Water Works Assn.* **1946**, *38*, 179–185. [[CrossRef](#)]
16. Ryzner, J.W. A new index for determining amount of calcium carbonate formed by water. *J. Am. Water Works Assn.* **1944**, *36*, 472–483. [[CrossRef](#)]
17. Alum, O.L.; Abugu, H.O.; Onwujiogu, V.C.; Ezugwu, A.L.; Egbueri, J.C.; Aralu, C.C.; Ucheana, I.A.; Okenwa, J.C.; Ezeofor, C.C.; Orjiocha, S.I.; et al. Characterization of the Hydrochemistry, Scaling and Corrosivity Tendencies, and Irrigation Suitability of the Water of the Rivers Karawa and Iyaji. *Sustainability* **2023**, *15*, 9366. [[CrossRef](#)]
18. Vasconcelos, H.C.; Fernández-Pérez, B.M.; González, S.; Souto, R.M.; Santana, J.J. Characterization of the Corrosive Action of Mineral Waters from Thermal Sources: A Case Study at Azores Archipelago, Portuga. *Water* **2015**, *7*, 3515–3530. [[CrossRef](#)]
19. Nakamura, Y.; Matsukawa, Y.; Okazaki, S.; Asakura, S. Proposal of a New Corrosion Index for Domestic Tap water in Japan. In Proceedings of the AMPP Annual Conference + Expo2023, Denver, CO, USA, 19–23 March 2023.
20. Sobue, K.; Magaino, S.; Sugahara, A.; Imai, H. The Effect of components Dissolved in Tap Water on Copper-corrosion Rate. *Zairyo-to-Kankyo* **2002**, *56*, 521–528.

21. So, Y.S.; Lim, J.M.; Kang, S.J.; Kim, W.C.; Kim, J.G. Derivation of Corrosion Depth Formula According to Corrosion Factors in District Heating Water through Regression Analysis. *Materials* **2023**, *16*, 3254. [[CrossRef](#)] [[PubMed](#)]
22. *JIS R 6253:2006*; Japanese Industrial Standards Committee (JISC), Waterproof Abrasive Papers. JISC: Tokyo, Japan, 2006; pp. 1–6.
23. Ministry of Health, Labour and Welfare of Japan. Available online: [https://www.mhlw.go.jp/english/policy/health/water\\_supply/4.html](https://www.mhlw.go.jp/english/policy/health/water_supply/4.html) (accessed on 6 December 2023).
24. *ASTM G31-72*; Standard Practice for Laboratory Immersion Corrosion Testing of Metals. ASTM International: West Conshohocken, PA, USA, 2004; pp. 1–8.
25. *NACE Standard TM-01-69*; Standard Guide for Laboratory Immersion Corrosion Testing of Metals. NACE International: Houston, TX, USA, 2000; pp. 1–9.
26. Uhlig, H.; Revie, R. *Corrosion and Corrosion Control*, 3rd ed.; Japanese Edition; Okamoto, G., Ed.; Sangyo Tosho: Tokyo, Japan, 1994; pp. 98–99.
27. Multiple Linear Regression (MLR) Definition, Formula, and Example. Available online: <https://www.investopedia.com/terms/m/mlr.asp> (accessed on 14 September 2023).
28. Variance Inflation Factor (VIF) A Measure of the Severity of Multicollinearity in Regression Analysis. Available online: <https://corporatefinanceinstitute.com/resources/data-science/variance-inflation-factor-vif/> (accessed on 14 September 2023).
29. Fujii, T. Water Quality and Corrosion of Metals. *J. Surf. Finish. Soc. Jpn.* **2000**, *51*, 134–140. [[CrossRef](#)]
30. Yuasa, M.; Sakai, Y.; Sekine, I.; Monozawa, N.; Wake, T.; Murata, H.; Shibara, Y.; Someya, S. Influence of Water Quality Factors on Stability Index of Water and Corrosion Rate of Steel. *Zairyo-to-Kankyo* **2000**, *49*, 568–571. [[CrossRef](#)]

**Disclaimer/Publisher’s Note:** The statements, opinions and data contained in all publications are solely those of the individual author(s) and contributor(s) and not of MDPI and/or the editor(s). MDPI and/or the editor(s) disclaim responsibility for any injury to people or property resulting from any ideas, methods, instructions or products referred to in the content.