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Atmospheric Corrosion Behavior of Carbon Steel by the Outdoor Exposure Test for 10 Years in Korea

Y. R. Yoo, S. H. Choi, and Y. S. Kim†

Materials Research Center for Energy and Clean Technology, School of Materials Science and Engineering,
Andong National University, 1375 Gyeongdong-ro, Andong, Gyeongbuk, 36729, Korea

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Steel was exposed in an atmospheric environment, and atmospheric environmental factors that include chloride, humidity, SO_2, NO_2 etc. induced the corrosion of steel. Corrosivity categories classified by SO_2 and chloride deposition rate were low, but those classified by TOW were high in the Korean Peninsula, and on these environmental categories, the corrosivity of atmospheres classified by corrosion rate in carbon steel was low medium, C2-C3, and medium, C3 for zinc, copper, and aluminum. This work performed the outdoor exposure test for 10 years at 14 areas in Korea and calculated the atmospheric corrosion rate of carbon steel. The atmospheric corrosion behavior of carbon steel is discussed based on the various corrosion factors. When the corrosion product forms on carbon steel by atmospheric corrosion, cracks may also be formed, and through these cracks, the environmental factors can penetrate into the interior of the product, detach some of the corrosion products and finally corrode locally. Thus, the maximum corrosion rate was about 7.3 times greater than the average corrosion rate. The color difference and glossiness of carbon steel by the 10 year-outdoor exposure tests are discussed based on the corrosion rate and the environmental factors.

Keywords: *Carbon steel, Atmospheric corrosion, Corrosion rate, Color difference, Glossiness*

1. Introduction

Carbon steel is a representative metallic material among metallic, ceramic, and polymeric materials, and has been widely used as a structural material, industrial piping, and in various fields, because it shows cost effectiveness and good mechanical properties [1]. Since steel is used for outdoor exposure, it shows weakness to atmospheric corrosion. If steel is exposed to an atmospheric environment, several environmental parameters may induce corrosion of the steel, and can then make a surface cracking and a reduction of thickness, and thus introduce a serious problem for industry.

When steel is exposed to an atmospheric environment, atmospheric environmental factors that include chloride, humidity, SO_2, and NO_2 induce its corrosion. If a chloride is deposited on the surface, the chloride facilitates the corrosion of steel. The chlorine particles and chloride can be transported via air ingression from sea water and salt-lake [2].

Atmospheric corrosion occurs through a thin electrolyte (*ca.* ~200 μm) containing several pollutants on the surface and the ambient temperature affects the corrosion [3-7]. The atmospheric corrosion environment can be classified into 4 kinds of categories of coastal, industrial, urban, and rural environments [8]. Coastal atmospheres are laden with fine particles of sea mist carried by the wind to settle on exposed surfaces as salt crystals. Industrial atmospheres are typically rich in various polluting gases that are composed mainly of sulfur compounds, such as sulfur dioxide, a precursor to acid rain, and nitrogen dioxide, the backbone of smog in modern cities. The corrosivity of urban and suburban environments is in great part related to the air quality in these environments, which can vary greatly from city to city, and from country to country. Typically, rural atmospheres are considered to be the most benign from a corrosion standpoint, since these atmospheres are free of industrial pollutants [9].

In the 1980s, three ambitious international programs on atmospheric corrosion (ISOCORRAG, ICP/UNECE and

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†Corresponding author: yikim@anu.ac.kr
Y. R. Yoo: Senior Researcher, S. H. Choi: Ph.D candidate, Y. S. Kim: Professor

MICAT), involving the participation of a total of 38 countries on four continents, Europe, America, Asia and Oceania, were launched [10]. Though each program has its own particular characteristics, the similarity of the basic methodologies used makes it possible to integrate the databases obtained in each case. In the damage function established for non-marine atmospheres, the great influence of the SO_2 content in the atmosphere was seen, as well as lesser effects by the meteorological parameters of Relative Humidity (RH) and Temperature (T). Both NaCl and SO_2 pollutants, in that order, are seen to be the most influential variables in marine atmospheres, along with a smaller impact of Time of Wetness (TOW) [10]. Based on these data, a simple classification scheme of five corrosivity classes was established for each metal [11], and these five corrosivity categories can be roughly translated into five outdoor situations listed in decreasing order of corrosivity, i.e., industrial, tropical marine, temperate marine, urban, and rural [12]. The present study showed the morphology of corrosion product film formed on mild steel after long-term atmospheric exposure (13 years) in five Spanish atmospheres of different types: rural, urban, industrial, mild marine, and severe marine [13] – the atmospheric corrosion rate was *ca.* 60 μm/y in severe marine atmosphere, *ca.* 50 μm/y in industrial atmosphere, *ca.* 30 μm/y in mild marine atmosphere, *ca.* 13 μm/y in urban atmosphere, and *ca.* 6 μm/y in rural atmosphere.

In Korea, atmospheric corrosion was first reported in Jeju Island based on the environmental index in 2003 [3], and the Corrosion Science Society of Korea has worked on the determination of the corrosion rate of various metals and alloys since 2006 [14,15]. Corrosivity categories classified by SO_2 and chloride deposition rate were low, but high classified by TOW in Korean Peninsula and on these environmental categories, corrosivity of atmospheres classified by corrosion rate in carbon steel was low medium, C2-C3, and medium, C3 for zinc, copper, and aluminum [15]. In Korea, Y. S. Kim et al. reported that the corrosion rate of carbon steel in rural area (Andong, southeastern Korea) was in the range (8.5 − 18.4) μm/y at the first year from the outdoor exposure test [11].

Therefore, this work performed the outdoor exposure test for 10 years at 14 areas in Korea, and calculated the atmospheric corrosion rate of carbon steel. The atmospheric corrosion behavior of carbon steel based on the various corrosion factors is discussed.

2. Experimental Methods

2.1 Outdoor Exposure Test

Table 1 shows the chemical composition of the carbon steel used in this work. The specimen was cut to a size of 200 mm × 200 mm × 1.54 mm and installed to have a slope of 45° under outdoor exposure conditions. It was installed on flat ground where the amount of sunlight was always constant, in airy conditions, and where almost no buildings existed. For outdoor exposure sites, 4 kinds of categories of coastal, industrial, urban, and rural environments were selected. The outdoor exposure sites were installed at a total 14 locations in Busan, Seosan, Gwangyang, Incheon, Pohang, Asan, Ansan, Goyang, Seoul, Suwon, Gwangju, Andong, Jochiwon, and Chuncheon [13].

After the outdoor exposure test, the method of KS D 9226 was used to measure the average corrosion rate of carbon steel specimens [15]. To remove the corrosion product, distilled water was added to 400 g ammonium citric acid to prepare 2,000 mL, and the specimen immersed at 80 °C for 20 min to remove the corrosion product. After the chemical cleaning, the weight of the carbon steel specimen was measured, and the average corrosion rate was obtained by substituting the weight change before and after outdoor exposure into the following equation [16].

$$\text{Corrosion rate, } \mu\text{m}/\text{y} = \frac{\Delta W}{A\rho T}$$

ΔW = Weight loss (mg), ρ_{Fe} = 7.86 (g/cm^3)

Table 1. Chemical composition of the carbon steel used in the test (wt%)

C	Si	Mn	P	S	Ni	Cu	Fe
0.452	0.155	0.678	0.0189	0.0029	0.01	0.008	Bal.

A = Exposed area (cm^2), T = Time (h)
*ΔW = a-b
a = Weight of the specimen before exposure test (mg)
b = Weight of the specimen by chemical cleaning after exposure test (mg)

After the outdoor exposure test, to measure the maximum corrosion rate of the carbon steel specimen, the corroded carbon steel specimen was cut to a size of 15 mm × 15 mm × 1.54 mm, ground with #2000 SiC paper, and then mirror-finished with 3 μm diamond paste. The maximum corrosion rate was obtained by measuring the minimum residual thickness excluding the corrosion product of the cross-section of the carbon steel specimen, using optical microscopy (AXIOTECH 100 HD, ZEISS, Oberkochen, Germany). To collect atmospheric corrosive environmental factors, the data from the Meteorological Administration of Korea were used to investigate the TOW, SO_2 concentration, and NO_2 concentration from 2009 to 2018, and then the average values were calculated and displayed.

2.2 Surface appearance and Corrosion product analysis

2.2.1 Chromaticity measurement

The chromaticity of carbon steel was measured by color difference meter (SP64, X-rite, USA). As a measurement standard for chromaticity, L*a*b* values determined by the Commission international de l'éclairage (CIE) were used [17]. Fig. 1 shows the spatial representation of the CIE LAB color space. In the L*a*b* color space, L* indicates lightness, and a* and b* are chromaticity coordinates. a* and b* are color directions: +a* is the red axis, –a' is the green axis, +b* is the yellow axis, and –b* is the blue axis [18]. For the chromaticity measurement method, after washing the exposed specimen in running water with a soft brush, the central part of the specimen was positioned on a finder, and then repeatedly measured three times to obtain the L*, a*, and b* values, using the average value. In addition, the values of ΔL*, Δa*, and Δb* were calculated to confirm the overall color change, and the color difference value ΔE*ab was calculated using the following equation [19].

$$\Delta E^{*}ab = \sqrt{(\Delta L^{*})^{2} + (\Delta a^{*})^{2} + (\Delta b^{*})^{2}}$$

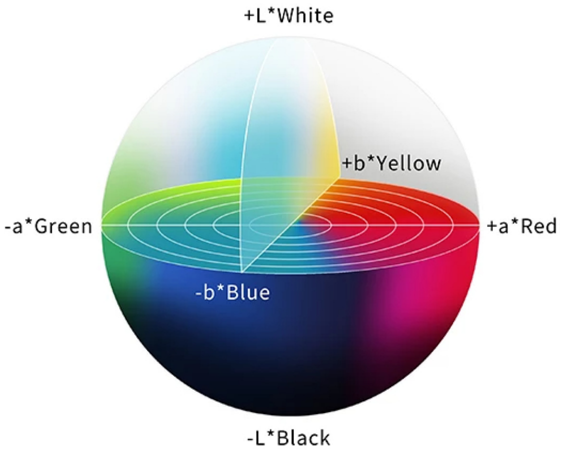


Fig. 1. CIE LAB color chart [18]

2.2.2 Gloss measurement

The gloss of carbon steel was measured by gloss meter (Micro-TRI-glass, BTK Ins., Germany). It was measured by using the method defined in the measurement method of KS L 2405 [20], and the international standard ASTM D 523 for gloss standard measurement [21]. Before measuring the gloss, foreign substances and corrosion products on the surface were removed by using a soft brush in flowing water, and then the average value was calculated by repeating measurements 5 times at a measuring angle of 60°.

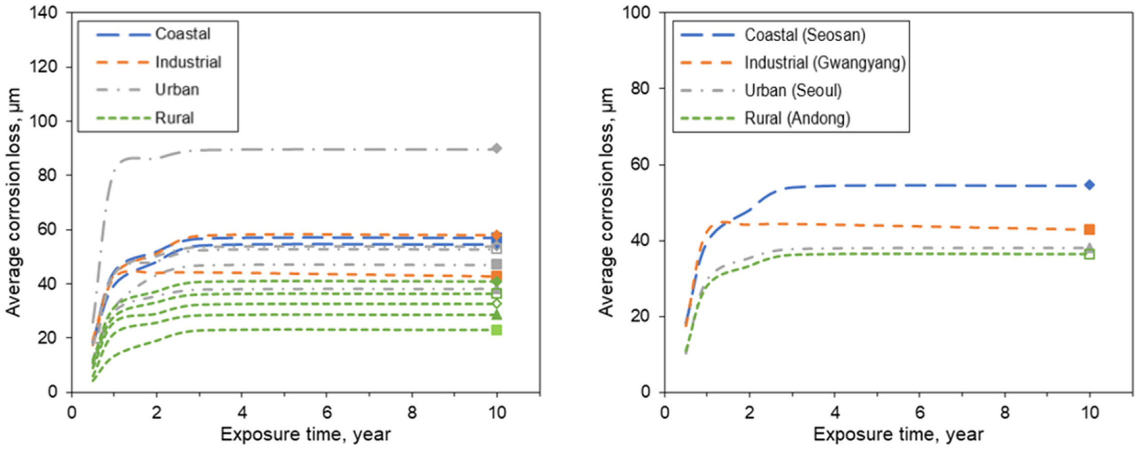
2.2.3 Cross-section analysis

After the outdoor exposure test, the specimen was cut to a size of 15 mm × 15 mm × 1.54 mm, the specimen was polished using #2000 SiC paper, and then mirror-finished with 3 μm diamond paste. The cross-sectional shape and composition were observed at ×400 magnification using SEM-EDS (MIRA3XMH, Tescan, Brno, Czech Republic) equipment and the contents of Fe and O in the cross-section were measured.

3. Results and Discussion

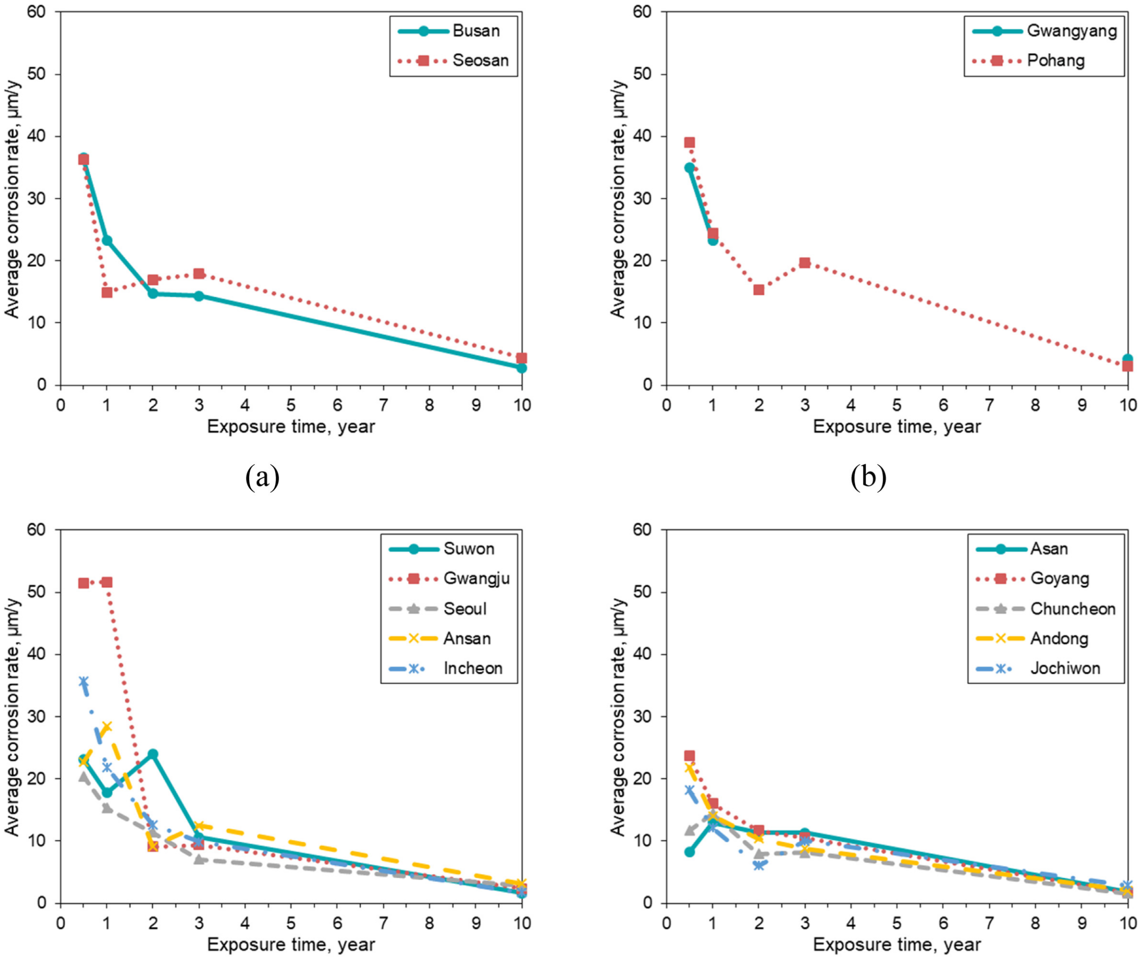
3.1 Atmospheric corrosion of carbon steel with the outdoor exposure time

Fig. 2a shows the average corrosion loss of carbon steel with the outdoor exposure time in Korea. The outdoor exposure tests were performed at 14 sites of coastal area (blue dashed line), industrial area (orange dashed line),



(a) (b)

Fig. 2. Average corrosion loss of carbon steel with the outdoor exposure time in Korea: (a) 4 atmospheric corrosion environments of 14 sites, and (b) four typical areas



(a) (b)

(c) (d)

Fig. 3. Average corrosion rate of carbon steel with outdoor exposure time in Korea for 10 years: (a) Coastal areas, (b) Industrial areas, (c) Urban areas, and (d) Rural areas

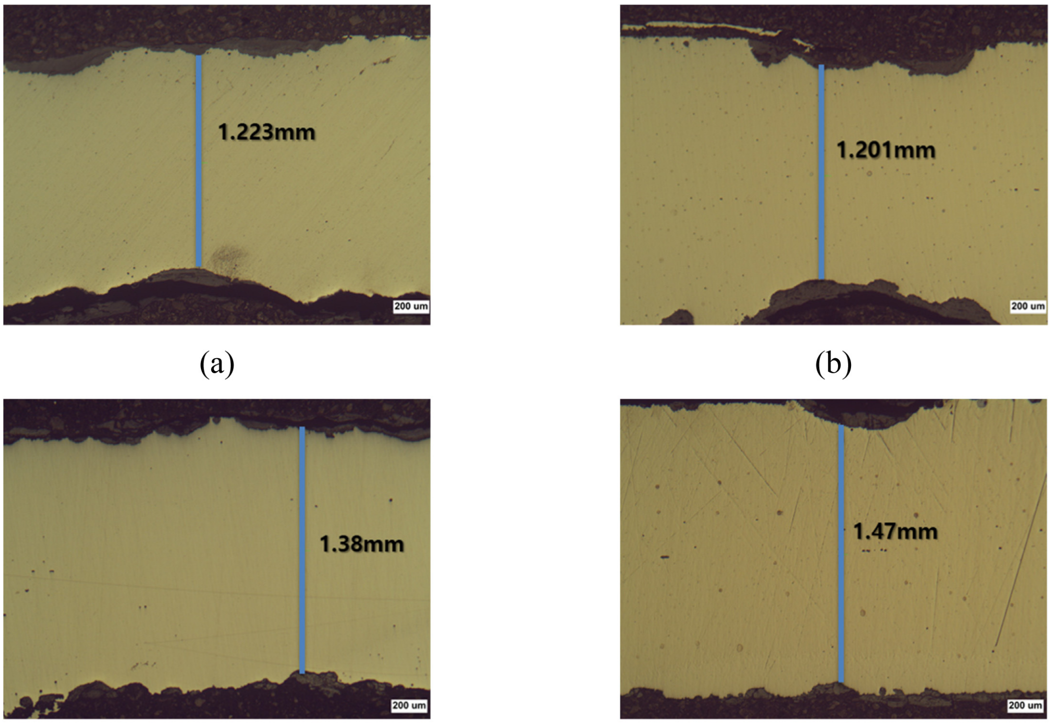
urban area (gray dashed & dotted line), and rural area (green dashed line). The figure shows that the average corrosion loss increased greatly till the 3 years exposure test, but the loss after 3 years was almost unchanged. Fig. 2b shows the average corrosion loss of 4 typical sites – Seosan (coastal area), Gwangyang (industrial and coastal area), Seoul (urban area), and Andong (rural area). The average corrosion loss of areas in the order greater to lesser was: coastal > industrial > urban > rural.

Fig. 3 reveals the average corrosion rate of carbon steel with the outdoor exposure time in Korea for 10 years. Fig. 3a shows the rate of coastal areas – Busan and Seosan; Fig. 3b shows the rate of industrial areas – Gwangyang and Pohang near the coastline; Fig. 3c shows the rate of urban areas – Suwon, Gwangju, Seoul, Ansan, and Incheon; and Fig. 3d shows the rate of rural areas – Asan, Goyang, Chuncheon, Andong, and Jochiwon. Regardless of the exposure area, the average corrosion rates in the early stage of the exposure test were very high; but as the exposure time increased, the average corrosion rate decreased. The high corrosion rate in Gwangyang and Pohang (Industrial area) in the early stage was due to airborne chloride, because both areas are near the coastline. In the case of urban area, except for Gwangju and Incheon, the others showed a relatively low corrosion rate. The high rate in the Incheon area was due to airborne chloride, because Incheon is near the coastline, but the high rate in the Gwangju area is very interesting, because Gwangju is an inland area. Among the exposure areas of the four types, the corrosion rate in the rural area was very low.

3.2 Relationship between atmospheric corrosion rate and environmental factors

Fig. 4 shows the cross sections of carbon steel after the outdoor exposure test for 10 years in Korea. Regardless of the exposure area, localized corrosion can be observed, even though there was general corrosion. In other word, the corrosion depth can differ, depending on the exposure areas. The maximum corrosion rate was determined by observing the thinned cross section using an optical microscope, and the average corrosion rate was obtained according to KS D ISO 9226 [15].

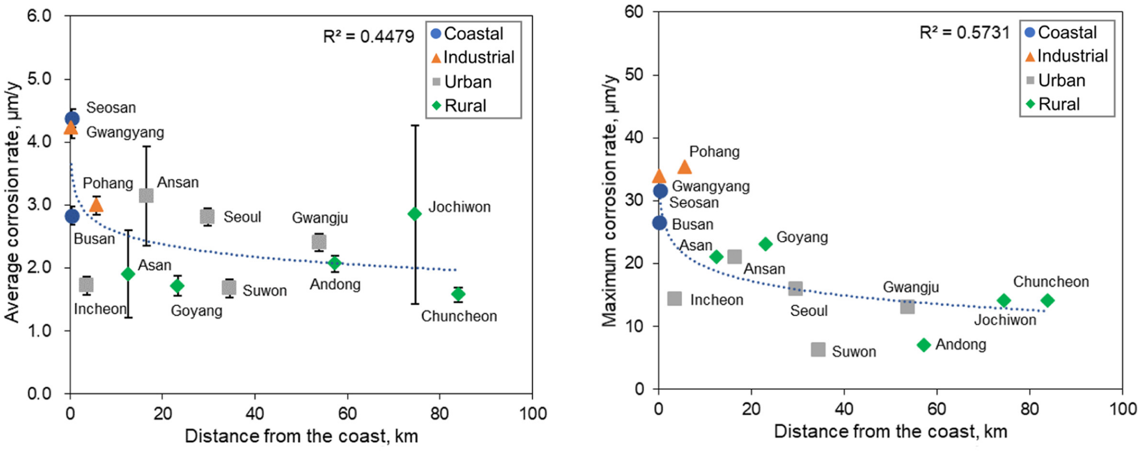
Fig. 5 shows the atmospheric corrosion rate of carbon



(a) (b)

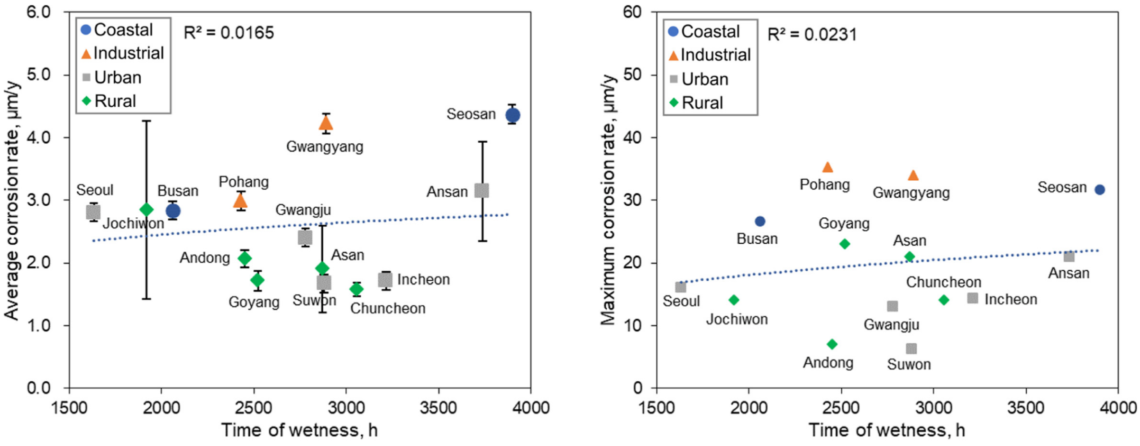
(c) (d)

Fig. 4. Localized corrosion of carbon steel exposed for 10 years in Korea: (a) Coastal area (Seosan), (b) Industrial area (Gwangyang), (c) Urban area (Seoul), and (d) Rural area (Andong)



(a) (b)

Fig. 5. Atmospheric corrosion rate of carbon steel exposed for 10 years in Korea with the distance from the coast: (a) Average corrosion rate, and (b) Maximum corrosion rate



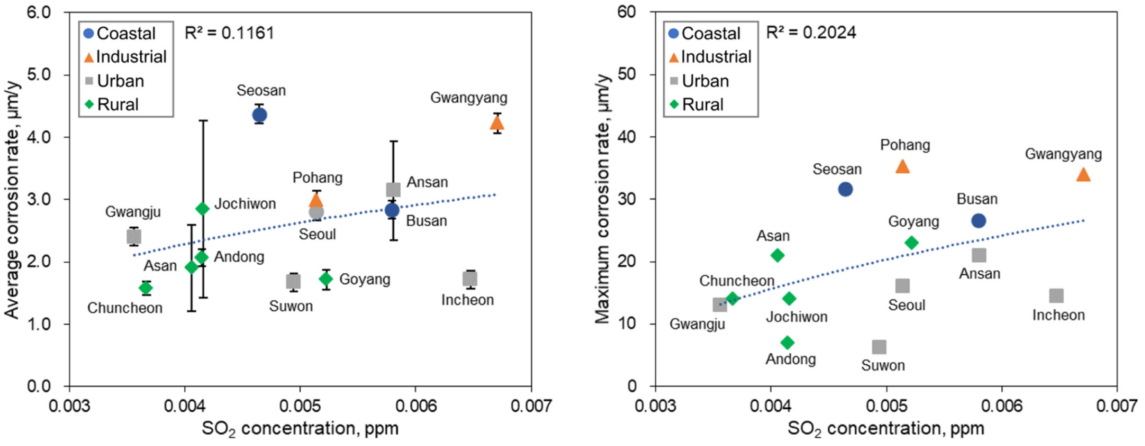
(a) (b)

Fig. 6. Relationship between the time of wetness and the atmospheric corrosion rate of carbon steel exposed for 10 years in Korea: (a) Average corrosion rate, and (b) Maximum corrosion rate

steel exposed for 10 years in Korea with the distance from the coast. Fig. 5a shows the average corrosion rate, while Fig. 5b shows the maximum corrosion rate. Even though there was a little fluctuation, the average corrosion and maximum corrosion rates increased as the exposure area was closer to the coast. The trend equation between the average corrosion rate and the distance from the coast was $y = -0.285\ln(x) + 3.2272$, and the trend equation between the maximum corrosion rate and the distance from the coast was $y = -3.334\ln(x) + 27.23$.

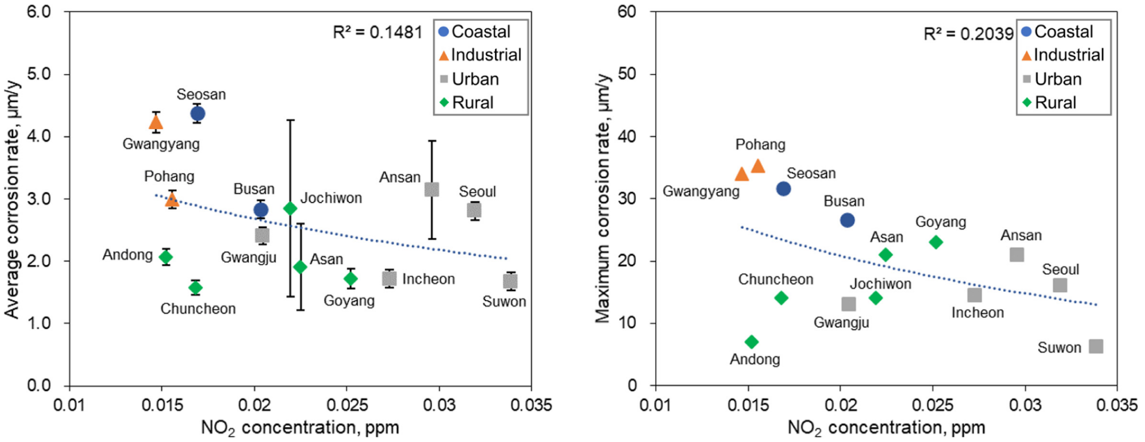
Fig. 6 depicts the relationship between the time of wetness and the atmospheric corrosion rate of carbon steel exposed for 10 years in Korea. With increasing time of wetness, the average corrosion rate and the maximum corrosion rate increased gradually, even though there was large fluctuation. The trend equation between the average corrosion rate and the time of wetness was $y = 0.4801\ln(x) - 1.1949$, while the trend equation between the maximum corrosion rate and the time of wetness was $y = 5.8805\ln(x) - 26.598$. However, it should be noted that the determination coefficients were very low.

Fig. 7 shows the relationship between SO_2 concentration, and the atmospheric corrosion rate of carbon steel exposed for 10 years in Korea. With



(a) (b)

Fig. 7. Relationship between SO_2 concentration and the atmospheric corrosion rate of carbon steel exposed for 10 years in Korea: (a) Average corrosion rate, and (b) Maximum corrosion rate



(a) (b)

Fig. 8. Relationship between NO_2 concentration and the atmospheric corrosion rate of carbon steel exposed for 10 years in Korea: (a) Average corrosion rate, and (b) Maximum corrosion rate

increasing SO_2 concentration, the average corrosion rate and the maximum corrosion rate increased gradually, even though there was large fluctuation. The trend equation between the average corrosion rate and the time of wetness was y = 1.5422ln(x) + 10.804, and the trend equation between the maximum corrosion rate and the time of wetness was y = 21.094ln(x) + 132.11. However, it should be noted that the determination coefficients were a little low.

Fig. 8 reveals the relationship between the NO_2 concentration and the atmospheric corrosion rate of carbon steel exposed for 10 years in Korea. As described above, although the environmental factors did increase the atmospheric corrosion rate, there was no relationship between NO_2 concentration and the atmospheric corrosion rate of carbon steel. However, it does not mean that NO_2 concentration did not affect the atmospheric corrosion of carbon steel.

Fig. 9 reveals the relationship between the average corrosion rate and the maximum corrosion rate obtained from the outdoor exposure test for 10 years in Korea. The trend equation between the maximum corrosion rate and the average corrosion rate was y = 7.2959x + 0.8785. That is, the maximum corrosion rate was about 7.3 times larger

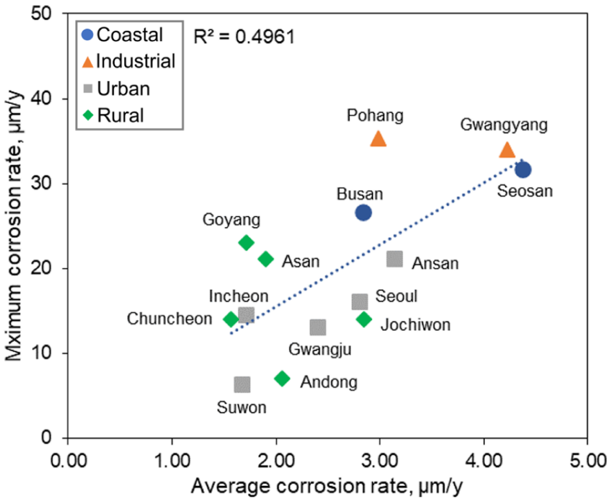


Fig. 9. Relationship between the average corrosion rate and the maximum corrosion rate obtained from the outdoor exposure test for 10 years in Korea

than the average corrosion rate, and this means that carbon steel may be corroded irregularly even with totally uniform exposure to corrosion.

Figs. 10a~d reveal the SEM images, while Figs. 10a'~d' depict the Fe-component mapping by EDS, and Figs. 10a"–d" show the O-component mapping by EDS. Fig. 10a shows the SEM for the coastal area (Seosan), Fig. 10b that for the industrial area (Gwangyang), Fig. 10c that for the urban area (Seoul), and Fig. 10d that for the rural area (Andong). The figures show that the corroded thickness was greater in the order coastal area > industrial area > urban area > rural area. Corrosion products formed in the coastal area and industrial area include some cracks. Most of the corrosion products consisted of iron oxide.

Based on the above test results, we could propose how

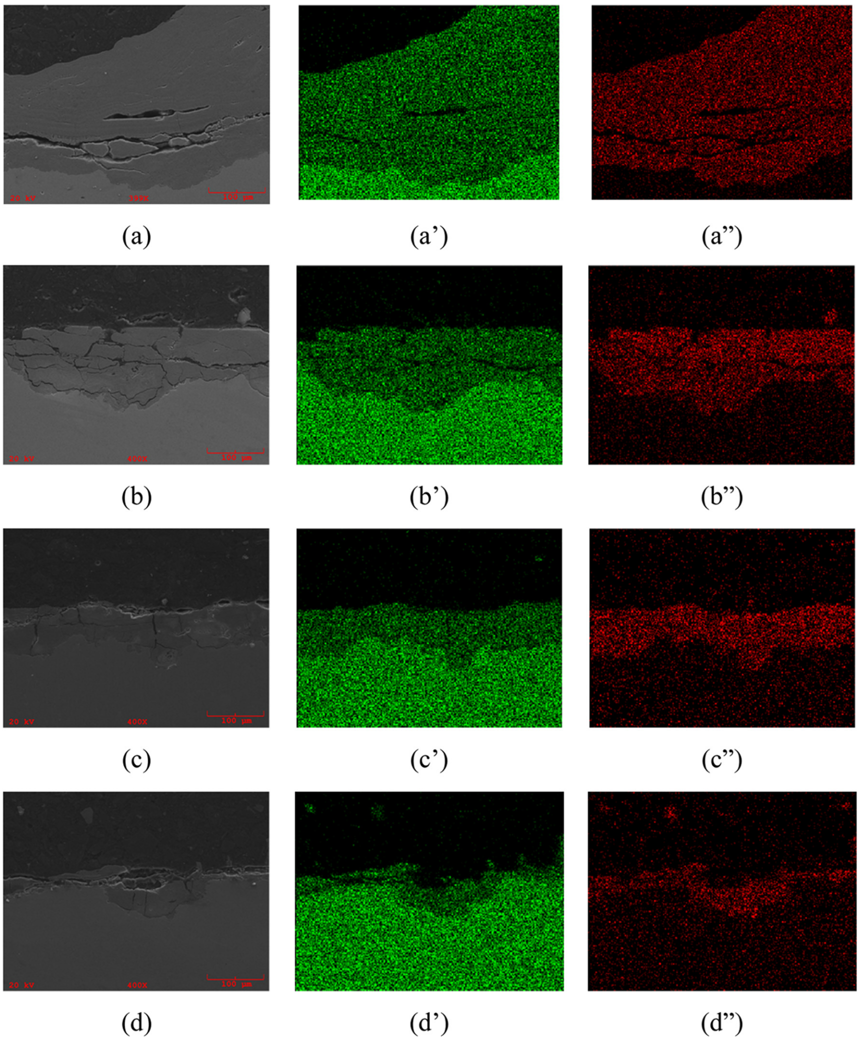


Fig. 10. (a~d) SEM images, (a'~d') Fe-component mapping by EDS, and (a"~d") O-component mapping by EDS: (a) Coastal area (Seosan), (b) Industrial area(Gwangyang), (c) Urban area (Seoul), and (d) Rural area (Andong)

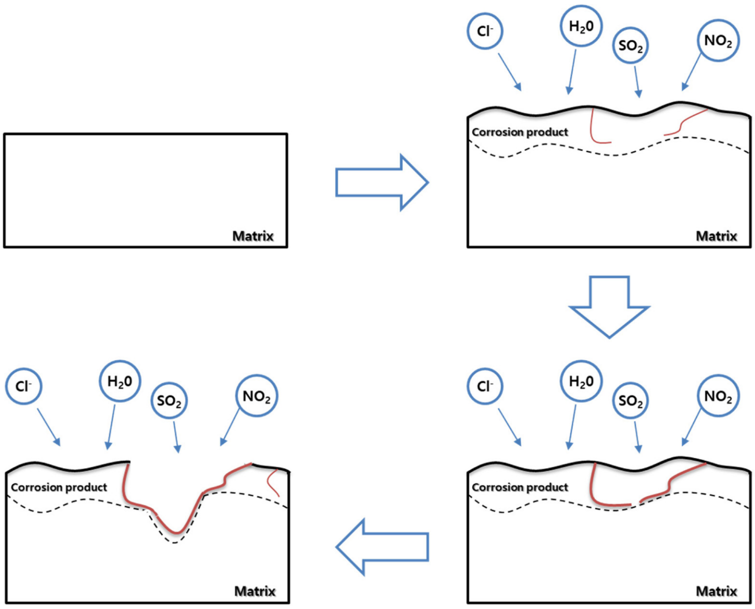


Fig. 11. Proposed mechanism of localized corrosion during atmospheric corrosion by outdoor exposure test

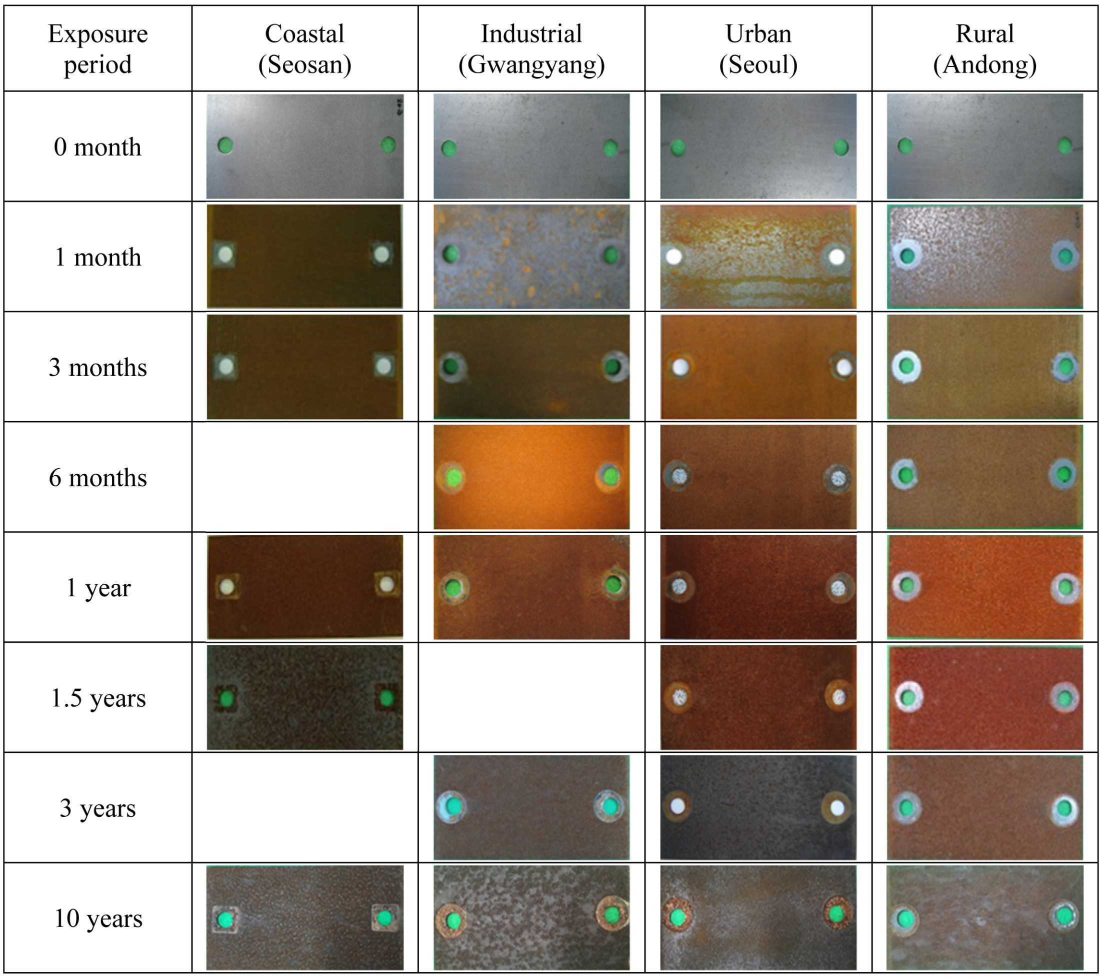


Fig. 12. Effect of the exposure time on the surface appearance of carbon steel by outdoor exposure test in 4 typical sites

localized corrosion can be possible during uniform exposure to corrosion. Fig. 11 shows the proposed mechanism of localized corrosion during atmospheric corrosion by outdoor exposure test. That is, when the corrosion product forms by atmospheric corrosion, cracks may also be formed, and through these cracks, the environmental factors can penetrate into the interior of the product, detach some of the corrosion products, and finally, corrode locally.

3.3 Surface appearance of carbon steel by the outdoor exposure test

Fig. 12 reveals the effect of exposure time on the surface appearance of carbon steel by outdoor exposure test in 4 typical sites – coastal area (Seosan), industrial area (Gwangyang), urban area (Seoul), and rural area (Andong). With increasing exposure time, the color of carbon steel changed from metal surface to red color. However, after (1.5 to 3) years, the color of corroded steel changed to black tone.

Fig. 13 shows the relationship between the average corrosion rate and the color difference of carbon steel exposed to the outdoors for 10 years in Korea. In the figure, the red dashed line indicates the color difference before the test. Regardless of the exposure sites, the color differences were increased. Note that there was no relationship between the corrosion rate of carbon steel and the color difference.

Fig. 14 shows the relationship between the blackening index (ΔL*) of carbon steel for 10 years exposed to the outdoors in Korea, and the (a) corrosion rate, (b) distance from the coast, (c) time of wetness, (d) SO_2 concentration, and (e) NO_2 concentration. In the figure, the red dashed line indicates the blackening index before the test. As can be seen, regardless of the exposure sites, the blackening was lowered. Note that there was no relationship between the corrosion rate and environmental factors, and the blackening index.

Fig. 15 shows the relationship between the redness index (Δa*) of carbon steel for 10 years exposed to the outdoors in Korea, and the (a) corrosion rate, (b) distance from the coast, (c) time of wetness, (d) SO_2 concentration, and (e) NO_2 concentration. In the figure, the red dashed line indicates the redness index before the test. As it can be seen, regardless of the exposure sites, the redness was increased. As the corrosion rate, time of wetness, and SO_2 concentration increased, the redness index was a little increased.

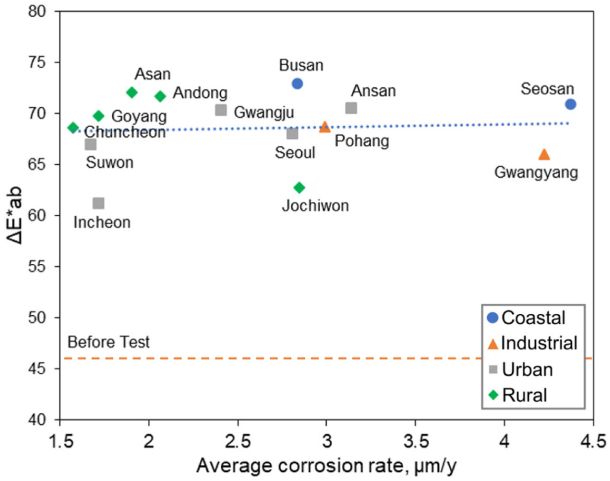


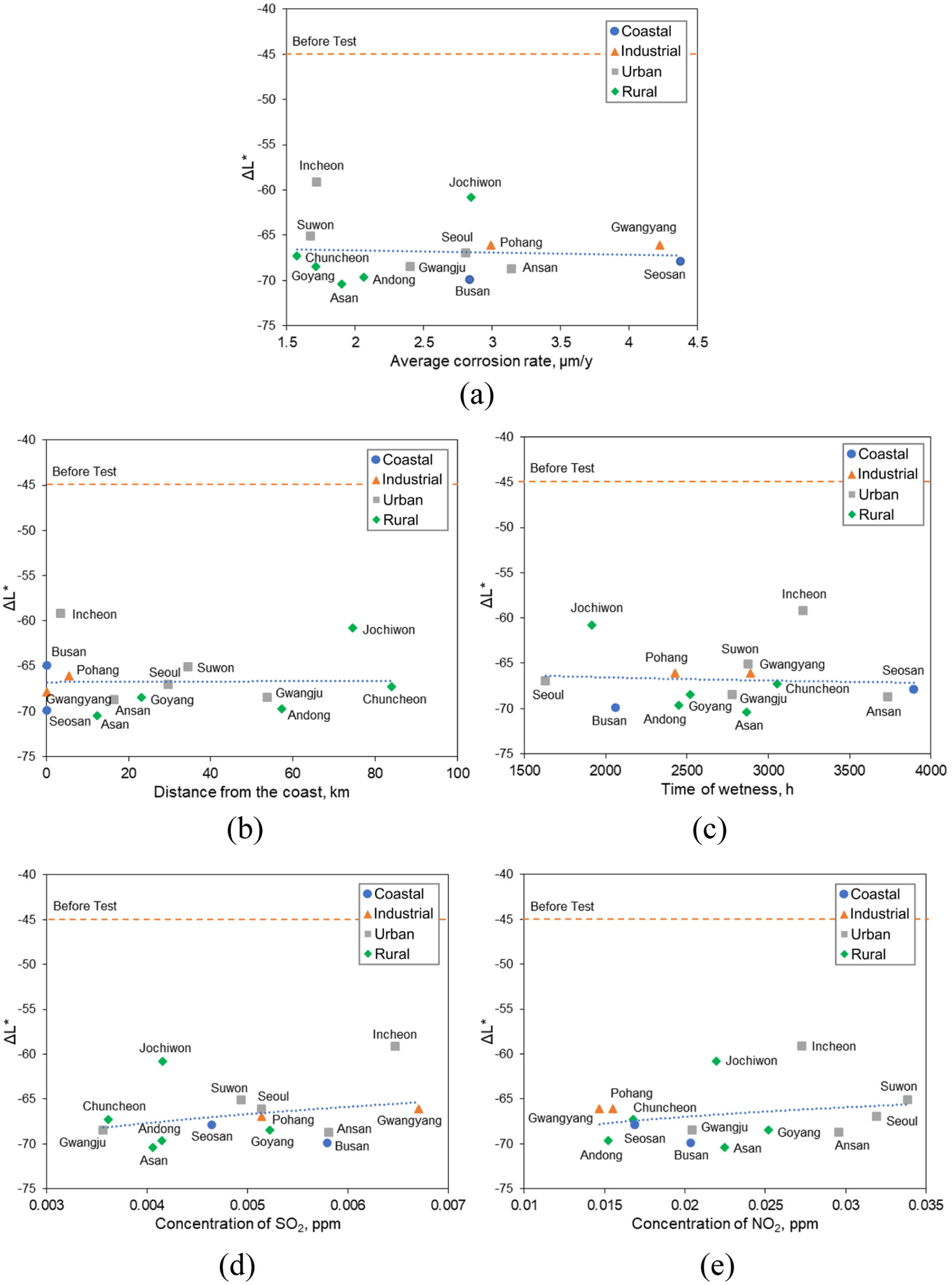
Fig. 13. Relationship between the average corrosion rate and the color difference of carbon steel exposed to the outdoors for 10 years in Korea

Fig. 16 shows the relationship between the yellowing index (Δb*) of carbon steel for 10 years exposed to the outdoors in Korea, and the (a) corrosion rate, (b) distance from the coast, (c) time of wetness, (d) SO_2 concentration, and (e) NO_2 concentration. In the figure, the red dashed line indicates the yellowing index before the test. As it can be seen, regardless of the exposure site, the yellowing was increased. As the corrosion rate, the time of wetness, and SO_2 concentration were increased, the yellowing index was a little increased.

Fig. 17 shows the relationship between the glossiness (ΔGloss) of carbon steel for 10 years exposed to the outdoors in Korea, and the (a) corrosion rate, (b) distance from the coast, (c) time of wetness, (d) SO_2 concentration, and (e) NO_2 concentration. In the figure, the red dashed line indicates the glossiness before the test. As it can be seen, regardless of the exposure sites, the glossiness was lowered. Note that there was no relationship between the corrosion rate and environmental factors, and the glossiness.

4. Conclusions

This work performed the outdoor exposure test of carbon steel for 10 years in Korea and the atmospheric



(a)

(b)

(c)

(d)

(e)

Fig. 14. Relationship between the blackening index (ΔL*) of carbon steel for 10 years exposed to the outdoors in Korea, and the (a) corrosion rate, (b) distance from the coast, (c) time of wetness, (d) SO_2 concentration, and (e) NO_2 concentration

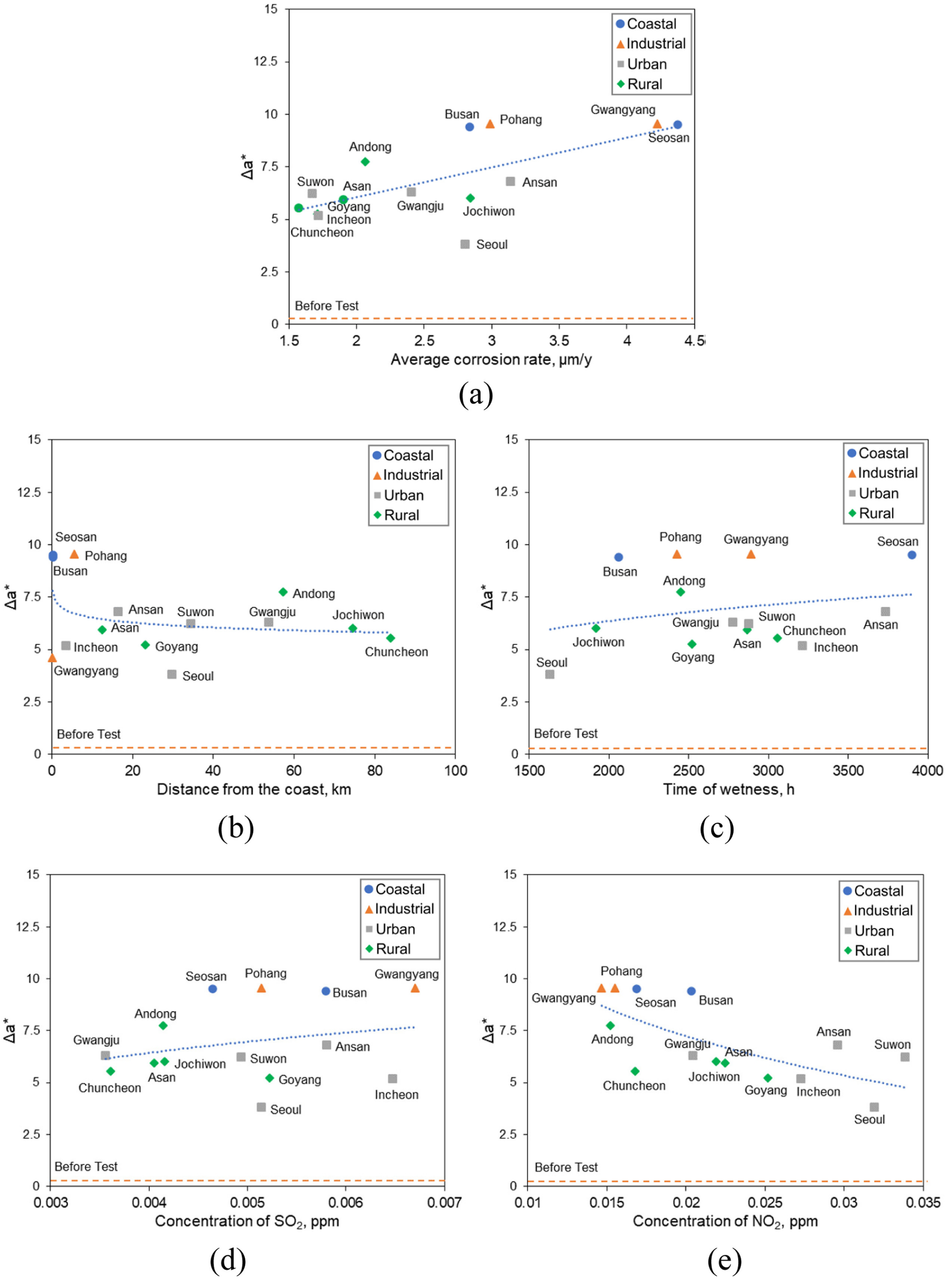
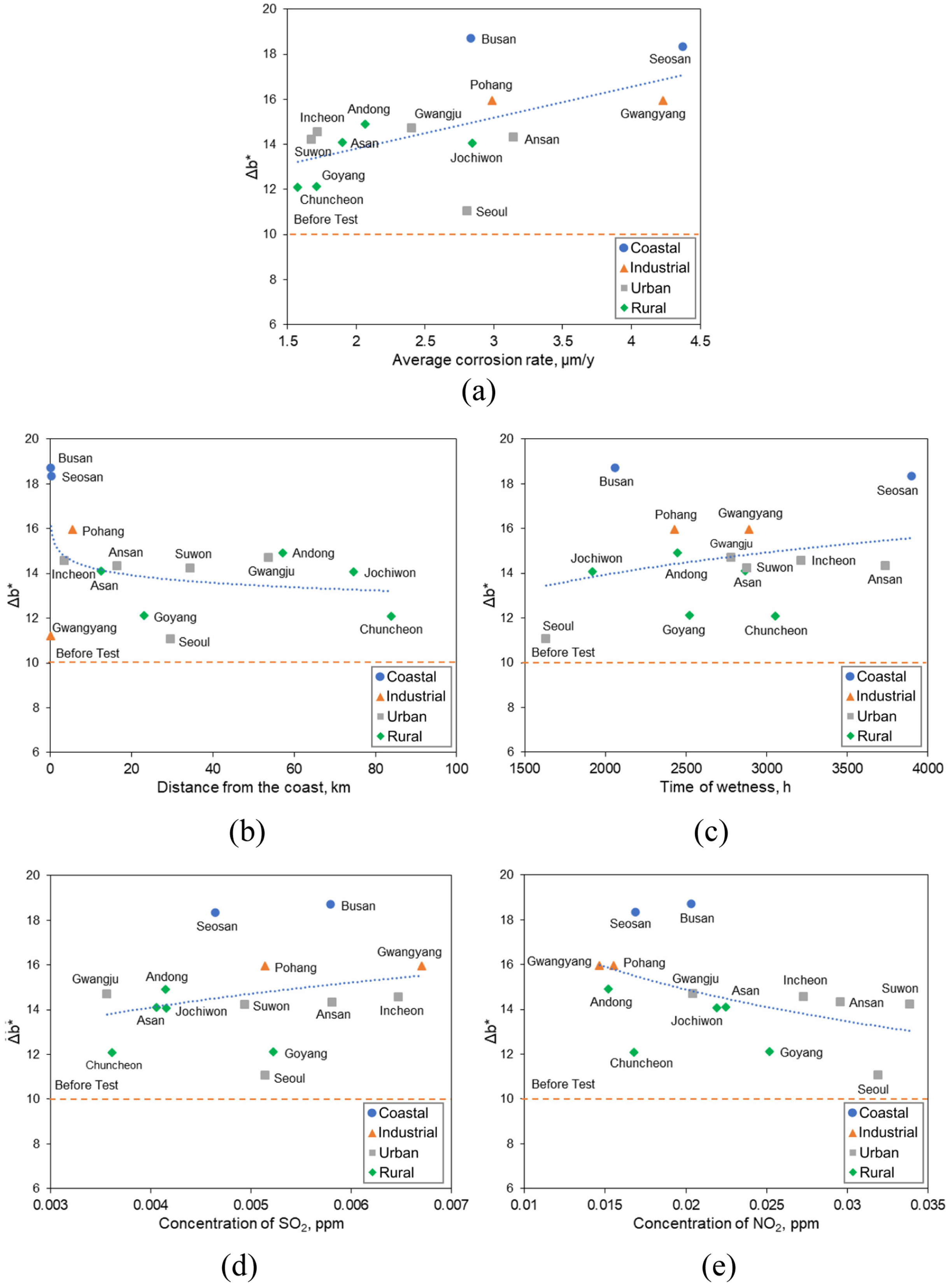


Fig. 15. Relationship between the redness index (Δa*) of carbon steel for 10 years exposed to the outdoors in Korea, and the (a) corrosion rate, (b) distance from the coast, (c) time of wetness, (d) SO_2 concentration, and (e) NO_2 concentration



(a)

(b)

(c)

(d)

(e)

Fig. 16. Relationship between the yellowing index (Δb*) of carbon steel for 10 years exposed to the outdoors in Korea, and the (a) corrosion rate, (b) distance from the coast, (c) time of wetness, (d) SO$_2$ concentration, and (e) NO$_2$ concentration

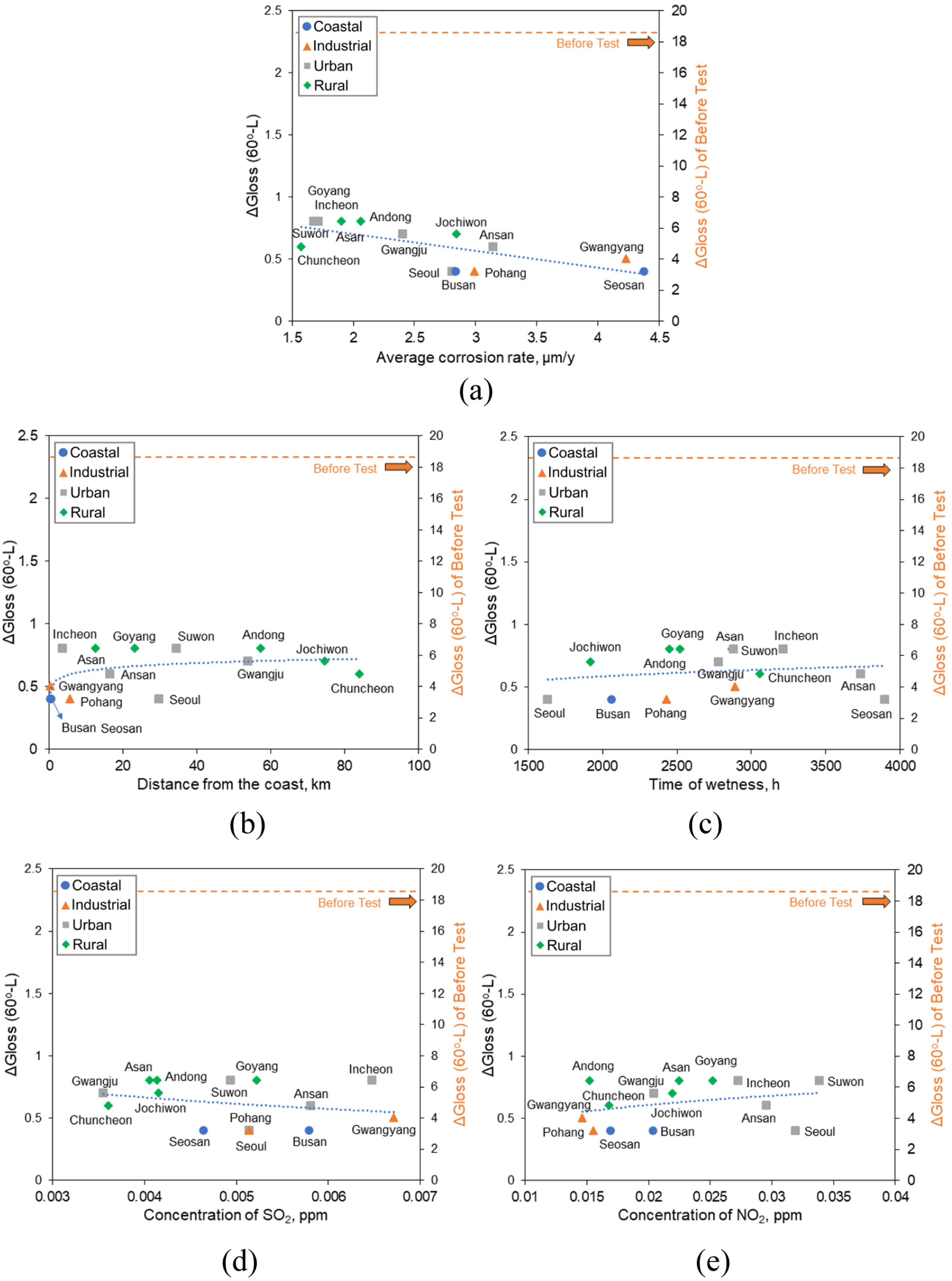


Fig. 17. Relationship between the glossiness (ΔGross) of carbon steel for 10 years exposed to the outdoors in Korea, and the (a) corrosion rate, (b) distance from the coast, (c) time of wetness, (d) SO$_2$ concentration, and (e) NO$_2$ concentration

corrosion rate was determined and the surface appearance was analyzed. Therefore, the following can be concluded:

1. The average corrosion rate in coastal area was obtained of from (2.83 to 4.23) μm/y, and the maximum corrosion rate was determined of from (26.6 to 31.7) μm/y. The average corrosion rate in industrial area was obtained of from (2.99 to 4.23) μm/y, and the maximum corrosion rate was determined of from (33.9 to 35.3) μm/y. The average corrosion rate in urban area was obtained of from (1.72 to 3.14) μm/y, and the maximum corrosion rate was determined of from (6.2 to 21.0) μm/y. The average corrosion rate in rural area was obtained of from (1.57 to 2.85) μm/y, and the maximum corrosion rate was determined of from (7.0 to 23.0) μm/y.
2. When the corrosion product forms on carbon steel by atmospheric corrosion, cracks may also be formed, and through these cracks, the environmental factors can penetrate into the interior of the product, detach some of the corrosion products, and finally corrode locally. Thus, the maximum corrosion rate was about 7.3 times greater than the average corrosion rate.
3. Regardless of the exposure sites, the color differences were increased. Note that there was no relationship between the corrosion rate of carbon steel and the color difference; the blackening was lowered, but there was no relationship between the corrosion rate and environmental factors, and the blackening index. However, when the corrosion rate, the time of wetness, and SO_2 concentration increased, the redness and yellowing indices were increased. On the other hand, regardless of the exposure sites, the glossiness was lowered and there was no relationship between the corrosion rate and environmental factors and the glossiness.

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