THE PREDICTION OF ATMOSPHERIC CORROSION FROM METEOROLOGICAL AND POLLUTION PARAMETERS—I. ANNUAL CORROSION

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Abstract—Worldwide atmospheric corrosion and environmental data, compiled in a comprehensive literature survey were statistically processed for establishing general corrosion damage functions for mild steel, zinc, copper and aluminium, in terms of simple meteorological and pollution parameters. The regression equations thus developed account for 83, 62, 59 and 41% of the variance in the annual corrosion data for zinc, steel, aluminium and copper, respectively. The goodness of the fits of data from rural atmospheres is considerably lower.

INTRODUCTION

Models for predicting the corrosion damage of metals in the atmosphere are useful for answering questions regarding the durability of metallic structures, determining the economic costs of damages associated with the degradation of materials, and acquiring knowledge about the effect of environmental variables on corrosion kinetics.

First-year atmospheric corrosion (A) is an important parameter in this respect as it allows not only the corrosiveness of a given atmosphere to be calibrated, but also long-term corrosion to be predicted. This latter use is clearly observed in the equation

$$C = At^n \tag{1}$$

according to which, corrosion C after t years depends directly on A, and follows a power law of time, where A and the exponent n are dependent on the type of metal and climatic parameters. The general validity of this equation and its reliability to predict long term corrosion behaviour has been demonstrated by many authors. For a rapid prediction of C, the possibility of estimating A and n from commonly available meteorological and pollution data is of great interest.

The specialized literature offers a large number of damage functions that relate A to environmental data, many of which, however, are of limited applicability as they were obtained by statistical processing of data involving a small variation in the meteorological parameters (chiefly because they corresponded to a rather small geographic area) and low-polluted sites. Ideally, the statistical analysis in question should be applied to data obtained under as diverse as possible meteorological and pollution conditions in order to establish universal damage functions. This was the chief aim of the work reported here.

The development of the aforementioned universal functions should therefore rely on worldwide corrosion and climatological data. Obtaining this information is

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obviously beyond the experimental possibilities of a research group, so it was necessary to supplement our data with those of many other groups. In this respect it should be noted that, despite the abundance of atmospheric corrosion data in the literature, many of them are not accompanied by simultaneous pollution and meteorological measurements, so they are of limited value.

Special mention should be made here of the effort of Benarie and Lipfert³ to develop universal corrosion functions in terms of atmospheric pollutants, meteorological parameters and the rain pH. For this purpose they used data from 72 corrosion test sites throughout the world. Our data base was considerably larger (e.g. 324 data obtained from 250 test sites were used to analyse annual corrosion of steel). Fortunately, the information on atmospheric corrosion reported in the last 6 years (i.e. after the aforementioned paper by Benarie and Lipfert was published) was available. Many of the new data were obtained from Spanish and Latin-American test sites. Had rain pH data been included, as in the Benarie and Lipfert study,³ the amount of usable data would have been much smaller, so that parameter was not used. One of the aims of this work was to check whether A and n in equation (1) could be estimated from widely available environmental parameters.

DATA COMPILATION

As much corrosion data as possible was collected from the literature in order to deal with a reasonably large set of data. The lack of sulphur dioxide and chloride pollution data from many corrosion test sites required a substantial portion of the initially available information to be discarded. As a whole, data were compiled from 76 references, 4–79 which are classified in Table 1 according to the countries for which information was reported.

Meteorological and pollution factors may vary widely throughout the atmospheric exposure period, so it is common practice to use annual averages. The meteorological parameters considered in the statistical analysis included the relative humidity (RH) and temperature (T) annual averages figures, as well as the number of rainy days per year (D). The wetness time ($t_{\rm w}$), which is generally believed to be associated with corrosion mechanisms was also included. Worldwide relative humidity, rainfall, rainy days and temperature data are available from climatological tables; however, reliable $t_{\rm w}$ data are not so commonplace. The problems faced in the direct determination of $t_{\rm w}$ often lead to its indirect estimate from RH or rainy days data. $^{3,41.80}$ Sulphur dioxide and chlorides are the most common corrosive agents in the atmosphere and they were the sole pollution variables included in this study.

Table 2 shows the high variability of the corrosion and environmental data used in the statistical treatment.

STATISTICAL ANALYSIS

Data were fitted both to a linear equation

$$A = a_1 + a_2 t_w + a_3 RH + a_4 D + a_5 T + a_6 S + a_7 Cl$$
 (2)

and to a logarithmic equation

$$\ln A = b_1 + b_2 \ln t_w + b_3 \ln RH + b_4 \ln D + b_5 \ln T + b_6 \ln S + b_7 \ln Cl$$
 (3)

where the a_i and b_i coefficients (i = 1 to 7) are constants, $A(\mu m)$ is the annual corrosion, t_w the wetness time (annual fraction), RH, D and T have the same

Table 1. References used to obtain information on the annual corrosion and countries for which information was reported

Country	References
Argentina	8, 40
Australia	66
Belgium	15, 71
Brazil	31, 78
Bulgaria	10, 11
Canada	41, 43, 74
Czechoslovakia	10, 11, 51, 54, 57, 62
Cuba	12, 13, 21, 22, 23, 24, 39, 46, 60, 61, 62, 69,
	70
France	15, 71
Germany	14, 55
Hungary	10, 11, 62
India	19, 53, 72, 73, 75
Italy	15, 71
Japan	67
New Zealand	20, 29
Nigeria	5, 52, 53
Norway	7, 44, 45, 56
Panama	38, 42, 76
Poland	79
Portugal	4, 27
Russia	10, 11, 62, 63
Spain	17, 25, 32, 33, 34, 35, 36, 64, 65, 68
South Africa	18
Sweden	44, 56, 57
Taiwan	37
U.K.	6, 30, 52, 53
U.S.A.	9, 16, 26, 30, 42, 47, 48, 49, 50, 58, 59, 77
Yugloslavia	28

meaning as above, $S \text{ (mg SO}_2 \text{ dm}^{-2} \text{ d}^{-1})$ is the annual average of SO_2 pollution and $Cl \text{ (mg } Cl^- \text{ dm}^{-2} \text{ d}^{-1})$ that of chloride.

The two equations above are quite simple. Probably, other combinations between the different variables may yield better fits, yet in this work using as simple relations as possible were aimed at. One possible way of improving the above linear equation is by introducing terms made up of products of two variables that are very likely to interact (e.g. $Cl \times S$, $T \times Cl$, $T \times S$, $t_w \times S$ and $t_w \times Cl$).

One major shortcoming of linear models is that they conform poorly to corrosion rate laws such as

$$C = k V^{l} \tag{4}$$

where C is the corrosion rate, V is the environmental variable concerned and k and l are two constants. This type of equation has been used to account for the effects of the pollution level⁸¹ and RH changes⁸² on the corrosion rate. In this case the logarithmic model is preferable. This model, however, does not allow for direct entering of zero pollution data and assumes that the effects of independent variables are multiplicative which is difficult to accept as a rule.

In addition to the simultaneous use of all the corrosion data, the present study included a separate treatment of data from rural atmospheres because some meteorological factors may act very differently in the presence or absence of pollutants, e.g. the lack of hygroscopic substances in a pure atmosphere probably results in thinner moisture films at a given RH value and in a less marked cleaning action of the rain. For the purpose of this work, rural atmospheres are defined as those with reported SO_2 and chlorides levels below $0.1 \text{ mg dm}^{-2} \text{ d}^{-1}$.

The goodness of fit of experimental data to a proposed model is often measured

Table 2. Characteristics of the corrosion and environmental data used in the statistical treatment

Material and number of data	Variable	Mean	SD	Smallest value	Largest value
Steel	corrosion (µm)	74.5	113.7	3.2	743
(N = 324)	RH (%)	73.8	6.7	38	87
`	$t_{\rm w}$ (annual fraction)	0.45	0.14	0.04	0.86
	number of rainy days per				
	year	129.7	39.6	16	231
	temperature (°C) chlorides	14.3	6.5	-1.2	29
	$(mg Cl^{-} dm^{-2} d^{-1})$	0.54	1.5	0	11.3
	$SO_2 (mg SO_2 dm^{-2} d^{-1})$	0.40	0.59	0	4.3
Zinc	corrosion (µm)	3.08	4.54	0.13	39.2
(N = 150)	RH (%)	72.2	7.1	37	85
	$t_{\rm w}$ (annual fraction) number of rainy days per	0.40	0.13	0.09	0.84
	year	120.7	44.4	25	231
	temperature (°C) chlorides	12.3	6.1	-1.2	27.4
	$(mg Cl^- dm^{-2} d^{-1})$	0.21	0.61	0	3.8
	$SO_2 (mg SO_2 dm^{-2} d^{-1})$	0.54	0.73	0	4.7
Copper	corrosion (µm)	2.01	1.78	0.25	12.1
(N = 119)	RH (%)	72.5	6.7	37	84
	$t_{\rm w}$ (annual fraction) number of rainy days per	0.39	0.13	0.14	0.84
	year	123.7	42.7	25	231
	temperature (°C) chlorides	12.6	5.3	-1.2	27.4
	$(mg Cl^- dm^{-2} d^{-1})$	0.22	0.56	0	3.8
	$SO_2 \text{ (mg } SO_2 \text{ dm}^{-2} \text{ d}^{-1})$	0.33	0.34	0	1.8
Aluminium	corrosion (µm)	0.60	0.85	0	3.7
(N = 91)	RH (%)	72.7	6.5	55	85
	$t_{\rm w}$ (annual fraction) number of rainy days per	0.43	0.15	0.14	0.86
	year	120.3	44.6	32	231
	temperature (°C) chlorides	12.6	6.4	0.5	27.0
	$(mg Cl^- dm^{-2} d^{-1})$	0.32	0.76	0	3.9
	$SO_2 (mg SO_2 dm^{-2} d^{-1})$	0.34	0.34	0	1.4

Equations $A = 132.4 \text{ Cl } (1 + 0.038 T - 1.96 t_w - 0.53 S + 74.6 t_w (1 + 1.07 S) - 6.3$		Remarks $A = \text{annual corrosion } (\mu \text{m}), t_{\text{w}} = \text{wetness}$ time (annual fraction), $T = \text{temperature}$	
A = 33.0 + 57.4 Cl + 26.6 S		Cl = chloride pollution annual average	
(R=0.73)	equation (6)	(mg Cl ⁻ dm ⁻² d ⁻¹), $R = \text{multiple correlation coefficient}$	

TABLE 3. RELATIONSHIPS BETWEEN THE ANNUAL CORROSION OF CARBON STEEL AND THE ENVIRONMENTAL PARAMETERS

by the statistical R^2 , i.e. the square of the correlation coefficient (R) between the observed values of the dependent variable and those predicted from the fitted line. Our selection of independent variables (three meteorological, two pollution variables and $t_{\rm w}$) was somewhat arbitrary. It is highly unlikely that all the relevant variables may have been considered, whether directly or indirectly. Also, some of the variables used may not be very important determinants of annual corrosion. Including all potentially relevant independent variables is as important as excluding irrelevant variables.

One way of assessing the relative importance of independent variables is to consider an increase in \mathbb{R}^2 when a variable is entered into an equation that already contains the other independent variables. A large change in \mathbb{R}^2 indicates that the new variable provides unique information about the dependent variable that is not available from the other independent variables in the equation. The procedure for selecting variables was the well-known stepwise technique, which involves the sequential introduction of all the independent variables starting from that one with the largest positive or negative correlation with the dependent variable. The value of the F statistic was used to determine whether this first variable and each of those successively introduced should remain in the regression equation or be excluded from it. In this way, only a few of the independent variables appeared to be important (equations in Tables 3–7).

The statistical analysis was performed with the aid of commercially available computer software. 83

Table 4. Relationships between the annual corrosion of zinc and the environmental parameters

Equations $A = 0.703 + 4.40 \text{ Cl}(1 + 0.11 T - 3.5 t_w) + 5.23 S(1 - 0.054 T)$		Remarks $A = \text{annual corrosion } (\mu \mathbf{m}),$ $t_{\mathbf{w}} = \text{wetness time (annual fraction)},$	
A = 0.785 + 5.01 Cl + 2.26 S ($R = 0.73$)	equation (8)	(mg SO ₂ dm ⁻² d ⁻¹), Cl = chloride pollution annual average (mg Cl dm ⁻² d ⁻¹), R = multiple correlation coefficient	

Equations		Remarks		
A = 0.94 + 0.11 Cl T + 2.31 S ($R = 0.64$)	(1 – 0.56 Cl) equation (9)	$A = \text{annual corrosion } (\mu \text{m}), T = \text{temperature annual average } (^{\circ}\text{C}), S = \text{SO}_2 \text{ pol-}$		
A = 1.03 + 1.45 Cl + 2.00 S	equation (3)	lution annual average (C_1 , $S = SO_2$ pollution annual average ($mg SO_2 dm^{-2} d^{-1}$), $Cl = chloride pollu-$		
(R = 0.60)	equation (10)	tion annual average (mg Cl ⁻ dm ⁻² d ⁻¹), R = multiple correlation coefficient		

Table 5. Relationships between the annual corrosion of copper and the environmental parameters

ANALYTICAL RESULTS

The statistical treatment of the whole data set by the linear model accounts for ca 50% of the variance in corrosion of steel and zinc and ca 40–45% in corrosion of copper and aluminium (equations 6, 8, 10 and 12 in Tables 3–6). The logarithmic model yields even poorer results for steel, zinc and copper, but accounts for 52% of the variance of aluminium corrosion.

The substantially improved fit of the linear model obtained by using binary interactions terms together with the main effect terms of the environmental variables (equations 5, 7, 9 and 11 in Tables 3–6) should be emphasized. In this way, the results for zinc accounted for 83% and for steel and aluminium ca 60% of their variances. The effect of the interaction (product) between the chloride pollution and the temperature is particularly important; the association between these two variables accounts for most of the variance in steel, zinc and copper corrosion.

Equations (5), (7), (9) and (11) represent the best least-squares fits obtained between the annual corrosion of mild steel, zinc, copper and aluminium, and the different independent variables which contribute significantly to the corrosion value. Together with these equations, those provided by the linear regression model in the absence of the interaction terms (equations 6, 8, 10 and 12) are presented. These latter fits were somewhat poorer, yet the equations have the advantage of showing the effect of variables in separate terms, so they lend to ready comparisons with other previously reported equations. Finally, it should be born in mind that great caution must be used in making corrosion predictions for values of the independent variables which are much larger or smaller than those used to derive these equations (Table 2).

Table 6. Relationships between the annual corrosion of aluminium and the environmental parameters

Equations $A = 0.13 + 2.19 \text{ Cl}(1 - 0.57 t_w - 0.015 T) + 0.57 S$ $(R = 0.77) \qquad \text{equation (11)}$ $A = 0.18 + 0.72 \text{ Cl} + 0.56 S$		Remarks $A = \text{annual corrosion } (\mu \text{m}), t_{\text{w}} = \text{wetness}$ time (annual fraction), $T = \text{temperature}$ annual average (°C), $S = \text{SO}_2$ pollution annual average (mg SO ₂ dm ⁻² d ⁻¹),	

Materials and number of data	Equations $A = 7.4 + 0.112 D$		Remarks $A = \text{annual corrosion } (\mu \mathbf{m}),$	
Steel				
(N=55)	(R=0.50)	equation (13)	D = number of rainy days per year, $T =$ temperature	
Zinc	$A = 0.28 + 0.52 10^{-3} D T$		annual average (°C),	
(n = 28)	(R=0.40)	equation (14)	RH = relative humidity annual average (%),	
Copper	$A = 0.41 + 0.57 10^{-3} D T$		R = multiple correlation co-	
(N=25)	(R = 0.56)	equation (15)	efficient	
Aluminium	A = 0.0049 RH	- 0.25		
(N = 23)	(R = 0.65)	equation (16)		

Table 7. Relationships between the annual corrosion and the environmental parameters for rural atmospheres

As can be seen from equations (5), (7), (9) and (11), the effect of the atmospheric pollutants appears to depend on the interaction with other variables as well. Thus, the effect of chlorides is multiplied by a factor that tends to increase with the temperature for steel, zinc and copper, and to decrease with the wetness time for steel, zinc and aluminium. For steel the effect of SO_2 tends to increase as t_w increases. Also, for steel and copper, the interaction between SO_2 and chlorides seems to moderate somewhat the joint effect of both pollutants.

Frequently, SO_2 and chlorides co-exist in the atmosphere. As a simplification, sometimes equivalent effects for the two pollutants are assumed, so the respective pollution levels in mg dm⁻² d⁻¹ can be added and introduced as a unique variable in the damage functions.³ Looking at the coefficients of SO_2 and chlorides in the regression equations (Tables 3–6), particularly the equations (6), (8), (10) and (12), that assumption could be roughly admitted for copper and aluminium. However, for mild steel and zinc the coefficient of chlorides is about twofold that of SO_2 .

It should be noted that the variance accounted for the regression equations decreases sharply if the statistical treatment is applied to rural atmospheres only (Table 7). The relative importance of independent variables changes from metal to metal. The most important variable for steel in this type of atmosphere was found to be D, although the linear regression equation accounted for only 25% of the variance. The behaviour of zinc and copper in rural atmospheres is similar to that of steel as regards the small fraction of variance accounted for the meteorological variables used in the statistical treatment. Inexplicably, RH accounts for up to 42% of the variance of aluminium in this type of atmosphere, equation (13).

THE GOODNESS OF THE FITS

As noted earlier, the environmental parameters considered in this work explain only partly the corrosion data. The regression equations for the model with interactions accounted for 83% of the variance in the best case (zinc) and 41% in the worst (copper). On the other hand, the unexplained variance was of the order of 50% or even higher for the other models. Such a high unexplained variance is attributable

not only to the oversimplification of the mathematical model, but also to the uncertainties inherent in the data and to the probable occurrence of other variables with marked effects on corrosion that were not considered in the statistical treatment.

The lack of quality in the corrosion data may arise from a number of factors.³ The uncertainty in the determination of t_w derived from problems posed by the use of wetness time monitors and the lack of precision of indirect measurements (e.g. in calculating the time at which RH exceeds a certain level) should be noted here. It is doubtful whether reported t_w values can be compared to one another as they were obtained by different procedures; meteorological parameters (RH, number of rainy days and temperature) are more reliable. Pollution by SO₂ and chlorides was also measured by a variety of methods, and expressed in different units. Conversions and equivalences are naturally prone to errors, so pollution data are liable to misinterpretations. Imprecisions in corrosion data often arise from deficient removal of corrosion products or metal attack by pickling baths, as well as from differences in the chemical composition, metallographic structure or internal stresses of metal specimens. Initial conditions of exposure (corrosion tests started in different months or seasons of the year), size, shape and orientation of the specimens, climatological changes throughout the exposure, and the number of replicates used in the corrosion tests are a few other factors that can influence the corrosion data. The lack of coincidence between years during which the metal was exposed and those in which the meteorological and pollution data were recorded, and between the locations of meteorological, pollution and corrosion measuring sites (effect of microclimates) are other potential sources of errors in this context. Fortunately, more reliable data are bound to become available in the near future as the results of such comprehensive projects on atmospheric corrosion as ISOCORRAG, 80 ICP/UNECE84 MICAT⁸⁵ are published. These three projects involve the joint effort of a number of researchers from many countries to unify operational procedures.

All of the above sources of imprecision by themselves might preclude the obtainment of better fits. However, the sources of errors are essentially the same irrespective of the metal, yet the regression equation for zinc accounted for 83% of the variance, whereas that for copper only accounted for 41%. Poor fits probably arise, therefore, not only from the aforementioned sources of imprecision, but also from the absence of certain (unidentified) variables from the regression equations which surely are specific to each metal, and from complex interactions between the variables that cannot be predicted by a linear model. One example of complex interactions involves the relative humidity, which, in addition to its effect on the wetting of the metal surface, plays a major role in the mechanisms whereby air pollutants take part in corrosion. ⁸⁶

Many effects have not been considered in these regression equations. For example, it is questionable whether the atmospheric corrosion of a given place depends entirely on the annual averages of environmental variables. It should also depend on the magnitude of the diurnal and seasonal changes in the meteorological and pollution parameters, as well as on the relative variations with time of some variables with respect to others. The frequency, duration and type of wetting and drying cycles to which metal surface is subjected may have a marked influence on the composition and morphology of the corrosion products layer. It has been shown⁸⁷ that frequent short wet periods followed by long dry periods give rise to the

formation of protective layers on steel. Sanyal and Singhamia⁸⁸ found that the lowest annual corrosion of steel was obtained when exposure had been initiated in the least aggressive months of the year.

Apart from sulphur dioxide and chlorides, other pollutants, including dust, smoke, nitrogen oxides, ozone, hydrogen peroxide, hydrocarbons, organic acids, etc., some of which are present in the atmosphere at extremely low concentrations, may play an important role in atmospheric corrosion. 80,89,90 Laboratory tests 91 have shown that SO₂ scarcely affects zinc corrosion at concentrations as high as 1 ppm; however, much lower concentrations of that gas produce appreciable corrosion in real atmospheres, possibly because of some synergistic effects of other pollutants or inorganic species. The supply rate of pollutant to the metal surface and solar radiation (sunshine hours) are another two parameters rarely considered in statistical relationships. The influence of low concentrations of SO₂ on corrosion rate has been shown to depend markedly on the flow velocity of the atmosphere; ⁹² consequently, the effect of this pollutant cannot be exclusively related to its concentration. Similarly, corrosion is affected by the velocity and duration of winds transporting the marine aerosol. 60,72 On the other hand, solar radiation rapidly increases the temperature of metal surfaces, so effective wetness time may decrease. This is probably the cause of the inverse relation occasionally found between corrosion and sunshine.²² In addition, solar radiation promotes photoelectrochemical activity in semiconducting corrosion product films. 93

Identifying every parameter that may significantly affect atmospheric corrosion of the different metals is a matter of scientific and technological interest and should thus be the subject of future research.

CONCLUSIONS

By using worldwide data compiled from an extensive literature survey, general damage functions for mild steel, zinc, copper and aluminium in terms of simple environmental parameters were obtained. The goodness of fit increased as a result of including the effect of some binary interactions between the independent variables in the statistical treatment. The product of the chloride level and the temperature was found to be a very important determinant of mild steel, zinc and copper corrosion. Also, the product of the SO_2 level and the wetness time was highly relevant to mild steel corrosion. In the best case, the regression equations account for up to 83% of the variance of zinc, 62% of mild steel, 59% of aluminium and only 41% of copper.

If the statistical analysis is restricted to the rural atmospheres only, then the unexplained variance is over 70% for mild steel, zinc and copper. This is interpreted in the sense that atmospheric corrosion is decisively affected by some unknown variables, different from the simple ones used in this analysis.

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