

# Exposure Programme on Atmospheric Corrosion Effects of Acidifying Pollutants in Tropical and Subtropical Climates

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**Abstract** Many national exposure programmes have been performed in tropical and subtropical climates during the last 50 years. However, ambitious programmes involving more than a few countries are scarce. In this paper a recently formed network of test sites is described involving 12 test sites in Asia (India, Vietnam, Thailand, Malaysia and China including Hong Kong) and four test sites in Africa (South Africa, Zambia and Zimbabwe). This effort is part of the 2001–2004 Swedish International Development Agency (SIDA) funded Programme on Regional Air

Pollution in Developing Countries (RAPIDC). Corrosion attack after one (2002–2003) year of exposure (carbon steel, zinc, copper, limestone and paint coated steel) are presented together with environmental data (SO<sub>2</sub>, NO<sub>2</sub>, HNO<sub>3</sub>, O<sub>3</sub>, particles, amount and pH of precipitation, temperature and relative humidity) for all the test sites. The obtained corrosion values are substantially higher than expected for limestone, higher than expected for carbon steel and lower than expected for zinc compared to values calculated using the best available dose–response functions.

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## 1 Introduction

Atmospheric corrosion research in tropical and sub-tropical climates started around 1945 and results are available from several individual countries, for example Australia, Brazil, China, Cuba, India, Manila, New Zealand, Nigeria, Panama, Papua New Guinea, Philippine Islands, South Africa, Taiwan, Singapore and Vietnam (Tidblad, Mikhailov, & Kucera, 2000). However, ambitious programmes involving more than a few countries are limited to the MICAT project (Morcillo, Almeida, Rosales, et al., 1998) which includes 12 Ibero-American countries, Spain and Portugal, and the ISOCORRAG programme with more than 50 sites located in Europe, Argentina, Canada, Japan, New Zealand and USA (Knotkova, 1993). In Asia, the programme co-ordinated from Australia and involving 13 sites in Australia, Philippines, Thailand and Vietnam (Cole, 2000) and the programme co-ordinated from Japan and involving 23 sites in Japan, China, and South Korea (Maeda et al., 2001) should be mentioned. In Africa, the long-term study of Callaghan (1991) should not be left unnoticed. These programmes either have an environmental characterisation limited to SO<sub>2</sub> as a main pollutant (Callaghan, 1991; Knotkova, 1993; Morcillo et al., 1998) or have relatively short exposure times (Cole, 2000; Maeda et al., 2001).

Partly in order to fill this gap the present project “RAPIDC/Corrosion” was initiated as a part of the 2001–2004 Swedish International Development Co-operation Agency (SIDA) funded Programme on Regional Air Pollution in Developing Countries (RAPIDC). The Programme was managed on SIDA’s behalf by the Stockholm Environment Institute (SEI). The corrosion project was co-ordinated by the Swedish Corrosion Institute (SCI). The objective of the present paper is to describe the corrosion project and to give results and conclusions after 1 year of exposure. Results will in the future be available after 2 and 4 years of exposure. Since 1 year of exposure is a relatively short time it should be stressed that the conclusions presented in this paper are preliminary in anticipation of the results after 2 and 4 years.

Another important aspect of the project, which is not described here, was transfer of knowledge on establishing test sites, exposing specimens, collection of environmental data, and methodology of developing dose–response functions using modelling and statistical treatment.

## 2 Materials and Methods

### 2.1 Rack and Test Sites

Each site was maintained by a dedicated partner who was responsible for the safety of the rack, the exposure/withdrawal of passive samplers, the collection of environmental data and the withdrawal of corrosion specimens. After exposure, the samples were returned to the Swedish Corrosion Institute for evaluation of corrosion attack. Table 1 shows a list of test sites including the responsible organisations and starting date of exposure. The network of test sites consists of six partners (12 sites) in Asia and three partners (4 sites) in Africa.

At each test site a rack was erected supporting the materials samples and passive samplers (Fig. 1). The rack should preferably be situated in an open space, preferably on a roof, and it contains three distinct elements: (1) flat metal/painted samples (a–d) and their support consisting of a wooden frame; (2) stone samples and their support consisting of a carousel (e); (3) passive samplers and their support and sheltering consisting of two metal discs (f).

### 2.2 Characterisation of the Environment

Passive sampling was performed on all sites for the gaseous pollutants SO<sub>2</sub>, NO<sub>2</sub>, O<sub>3</sub> and HNO<sub>3</sub> (Ferm, De Santis, & Varotsos, 2005), and for particulate matter (Ferm, Watt, O’Hanlon, De Santis, & Varotsos, 2006). Sampling was performed on a bi-monthly basis i.e., samplers were exchanged each second month. The total sampling period was one year making in total six bi-monthly sampling periods. The starting date of the first sampling period varied from site to site and coincided with the starting date of the exposure of specimens given in Table 1. All exposure periods for passive samplers were consecutive so that the end of a sampling period marked the start of a new sampling period.

**Table 1** List of test sites including country, responsible organisation and starting date of the exposure (year 2002)

Country	Test site name	Responsible organisation	Start
India	Bhubaneswar-u	Regional Research Laboratory	25 May
India	Bhubaneswar-r	Regional Research Laboratory	25 May
Thailand	Bangkok	Thailand Institute of Scientific and Technological Research	10 June
Thailand	Phrapradaeng	Thailand Institute of Scientific and Technological Research	11 June
Vietnam	Hanoi	Institute of Materials Science	23 July
Vietnam	Ho Chi Minh	Ho Chi Minh Branch of the Institute of Materials Science	26 July
Vietnam	Mytho	Ho Chi Minh Branch of the Institute of Materials Science	27 July
China	Chongqing	Chongqing Institute of Environmental Science and Monitoring	20 July
China	Tie Shan Ping	Chongqing Institute of Environmental Science and Monitoring	20 July
China	Hong Kong	Hong Kong Environmental Protection Department	18 July
Malaysia	Kuala Lumpur	Malaysian Meteorological Service	13 June
Malaysia	Tanah Rata	Malaysian Meteorological Service	14 June
South Africa	Johannesburg	CSIR / Materials and Manufacturing	21 August
Zambia	Kitwe	University of Zambia	27 August
Zambia	Magoye	University of Zambia	25 August
Zimbabwe	Harare	University of Zimbabwe	30 August

Complementary data on temperature, relative humidity, amount of rain and its pH were collected by the partners at a nearby environmental station. These data were reported to the Swedish Corrosion Institute on a monthly basis.

### 2.3 Sample Preparation and Evaluation of Corrosion Attack

For each of the materials and exposure periods, a set of three identical samples was prepared and exposed and these are referred to as the ‘triplicates.’ At each site a total of nine samples were exposed in the summer of 2002 for each material and this means that there are three sets of triplicates and that in total the samples are sufficient for three exposure periods. At present date withdrawals have been made after 1 and 2 years of exposure and a withdrawal after 4 years of exposure is planned in the summer of 2006. The samples exposed for 2 years are currently evaluated.

All flat samples, carbon steel (Dc 04, SS – EN 10 130), zinc (Z1 – DIN EN 1179), copper (Cu DHP, SS 5015) and painted steel were cut to dimensions  $100 \times 150 \text{ mm}^2$  as suggested by ISO 9226. The thickness of all flat samples was 1 mm except for steel, which had a thickness of 2 mm. Carbon steel, zinc and copper were degreased and weighed prior to exposure. Steel was painted with two layers of alkyd (90  $\mu\text{m}$ ): (1) Conseal Primer 50  $\mu\text{m}$ , fast drying alkyd based primer; (2) Ultra Topcoat 40  $\mu\text{m}$ , quick drying and glossy acrylic



**Fig. 1** Rack erected at a roof of a building in the urban site in Bhubaneswar: **a** carbon steel samples; **b** zinc samples; **c** copper samples; **d** painted steel samples; **e** limestone samples on a carousel; **f** passive samplers for gases and particulates fastened under circular discs for rain protection

modified alkyd topcoat and then edge protected using Vinyguard Silvergrey 88 from Jotun, Norway.

After exposure the visual impression of all flat samples was recorded by photography. The corrosion attack of the metal samples was evaluated with 10 min. consecutive pickling using Clarkes solution; 20 g  $\text{Sb}_2\text{O}_3$  and 60 g  $\text{SnCl}_2 \cdot 2\text{H}_2\text{O}$  and 1,000 ml concentrated HCl ( $\rho = 1.19 \text{ g ml}^{-1}$ ) for steel, 250 g glycine ( $\text{NH}_2\text{CH}_2\text{COOH}$ ) and distilled water to make 1,000 ml, saturated solution for zinc and 50 g amidosulfonic acid (sulfamic acid) and distilled water to make 1,000 ml for copper. Painted steel was evaluated by visual examination of the spread of corrosion attack in both directions from the 1 mm cut but expressed as the average spread in one direction following ASTM D 1654-79a.

Portland limestone specimens of dimensions  $50 \times 50 \times 8 \text{ mm}^3$  were obtained from the Building Research Establishment Ltd, United Kingdom, where also the corrosion attack was evaluated as mass change during exposure. The mass change was then recalculated to surface recession.

### 3 Results and Discussion

As mentioned in the introduction, 1 year of exposure is a relatively short time and therefore it is not

worthwhile at this stage to perform a detailed statistical analysis and development of dose–response functions. This analysis will be performed when data after 2 and 4 years of exposure are available. The focus is instead on presenting the data set and its characteristics and to make an estimate on the magnitude of the effects compared to those predicted by existing dose–response functions.

#### 3.1 Environmental Data

The environmental data are given in Table 2. Compared to European values the values in Table 2 are similar except for temperature, amount of precipitation and  $\text{SO}_2$  concentration, where the values are generally higher.

The  $\text{SO}_2$  values are lower than  $20 \mu\text{g m}^{-3}$  except for four extreme sites: Phrapradaeng, Chongqing, Tie Shan Ping and Kitwe. Worth noting is that the site Tie Shan Ping is a *rural* site situated close to Chongqing. The site Kitwe is located in the copper belt area in the northern part of Zambia. Other extreme sites worth mentioning are those in Malaysia: Kuala Lumpur and Tanah Rata. Kuala Lumpur has the highest  $\text{HNO}_3$  values in the network probably due to a combination of the high  $\text{NO}_2$  emissions, the high temperature, and the humid conditions. Tanah Rata is the cleanest site in the network. It has the same precipitation level as

**Table 2** Environmental data including temperature ( $T$ ), relative humidity (Rh), precipitation (Rain and pH), gaseous pollutants ( $\text{HNO}_3$ ,  $\text{SO}_2$ ,  $\text{NO}_2$  and  $\text{O}_3$ ) and particulate deposition (mass  $\text{Cl}^-$ ,  $\text{NO}_3^-$  and  $\text{SO}_4^{2-}$ )

Name	Temperature	Relative humidity	Rain	pH	$\text{HNO}_3$	$\text{SO}_2$	$\text{NO}_2$	$\text{O}_3$	mass	$\text{Cl}^-$	$\text{NO}_3^-$	$\text{SO}_4^{2-}$
Unit	$^\circ\text{C}$	%	mm		$\mu\text{g m}^{-3}$				$\mu\text{g cm}^{-2} \text{ month}^{-1}$			
Bhubaneswar–u	26.5	69	425	6.0	1.3	4	11	63	164	5.8	2.38	3.1
Bhubaneswar–r	26.5	69	425	6.0	1.0	3	5	63	70	1.7	1.02	1.4
Bangkok	29.3	76	1,371	6.8	2.3	11	39	38	35	0.3	2.18	1.6
Phrapradaeng	29.3	73	1,335	6.2	1.5	59	24	54	64	0.9	1.88	6.6
Hanoi	24.7	79	1,556	5.8	0.8	15	18	49	57	0.4	1.08	3.4
Ho Chi Minh	28.3	74	1,441	6.2	0.9	21	18	47	39	0.4	0.82	2.4
Mytho	27.0	81	1,222	6.4	0.3	2	9	36	62	2.9	0.62	1.3
Chongqing	18.5	70	1,162	4.5	1.3	99	45	52	162	0.7	1.46	18.5
Tie Shan Ping	18.5	90	1,133	4.2	1.8	51	10	71	61	0.2	1.25	7.2
Hong Kong	22.9	78	2,092	4.6	1.8	16	50	31	36	0.8	1.98	1.6
Kuala Lumpur	28.0	78	2,776	4.3	3.8	12	47	42	26	0.2	1.42	1.0
Tanah Rata	18.1	91	2,433	5.1	0.1	0	1	35	10	0.1	0.25	0.3
Johannesburg	17.2	78	417	4.8	2.1	18	28	51	21	0.1	0.62	0.7
Kitwe	22.6	58	1,083	4.7	0.9	92	11	72	64	0.2	0.52	6.5
Magoye	22.2	62	826	7.0	0.5	0	2	53	24	0.1	0.20	0.1
Harare	18.9	63	798	6.6	0.7	16	15	65	31	0.1	0.36	1.3

Kuala Lumpur but a much lower temperature and is situated in the Cameron highlands.

### 3.2 Corrosion Data

The corrosion data are given in Table 3. The analysis of painted steel and limestone are non-destructive while the analysis (pickling) of the metals are destructive and this is the reason why one of each of the triplicate metal samples (no. 3) have been saved for further analysis by other methods. As can be seen from Table 3 the difference between no. 1 and no. 2 for the metals is generally very low, in the range of 5%.

Regarding zinc one should note the slightly higher corrosion values for zinc at the Tie Shan Ping test site compared to Chongqing, which has a much higher SO<sub>2</sub> value, and also the very high values observed in Kitwe. For limestone, the high values in Johannesburg deserves to be mentioned, considering the pollution situation.

### 3.3 Correlation Between Environmental and Corrosion Data

Table 4 shows the rank correlations between the corrosion and selected environmental parameters. The

environmental parameter that has the highest correlation to most of the parameters is the amount of SO<sub>4</sub><sup>2-</sup> analysed from the particulate deposited matter. It should be mentioned that SO<sub>4</sub><sup>2-</sup> is not necessarily deposited as sulphate but may also be the reaction product of other particulate deposits, probably CaCO<sub>3</sub>, and gaseous SO<sub>2</sub>.

Other parameters with high correlation are SO<sub>2</sub> for carbon steel, pH for zinc and SO<sub>2</sub>, pH and HNO<sub>3</sub> for limestone. Nitric acid is a highly corrosive gas, even compared to SO<sub>2</sub>, as has been shown in recent laboratory experiments for copper (Samie, Tidblad, Kucera, & Leygraf, 2005).

### 3.4 Comparison of Observed and Predicted Data

Since it is not possible to develop dose–response functions based on the data presented in Table 2 and 3 due to the short exposure time it is instead worthwhile to use existing dose–response functions and compare the predicted values with the experimental values. Functions have recently been developed for the multi-pollutant situation within the EU 5FP project Model for multi-pollutant impact and assessment of threshold levels for cultural heritage (MULTI-ASSESS). These functions are also pre-

**Table 3** Corrosion attack after 1 year of exposure (2002–2003) on painted steel (spread from cut), carbon steel, zinc and copper (mass loss) and limestone (surface recession)

Name	Painted steel			Carbon steel		Zinc		Copper		Limestone		
	mm			g m <sup>-2</sup>						µm		
Sample	No. 1	No. 2	No. 3	No. 1	No. 2	No. 1	No. 2	No. 1	No. 2	No. 1	No. 2	No. 3
Bhubaneswar-u	0.6	0.6	0.7	157	157	4.2	4.3	8.4	8.1	13.2	14.3	12.5
Bhubaneswar-r	0.6	0.8	0.7	157	155	3.4	3.7	12.3	11.5	7.7	8.9	11.5
Bangkok	0.8	0.6	0.7	116	115	4.3	4.8	14.3	15.6	13.2	13.9	15.2
Phrapradaeng	0.6	0.5	0.6	280	282	5.5	5.9	16.8	17.4	28.9	3.0	17.3
Hanoi	1.0	1.4	1.1	181	182	5.9	6.2	5.5	5.2	23.1	5.4	21.8
Ho Chi Minh	0.8	0.9	0.8	162	165	6.4	7.3	7.7	8.3	5.3	7.6	7.1
Mytho	1.2	0.9	1.0	166	167	4.4	4.3	11.3	12.8	9.3	7.4	11.5
Chongqing	4.0	5.3	4.4	789	776	9.1	9.0	24.6	23.7	17.5	38.7	37.4
Tie Shan Ping	4.0	4.2	3.1	490	494	10.8	12.4	17.7	18.0	30.1	29.4	33.8
Hong Kong	0.7	0.7	0.7	148	153	6.4	6.4	6.6	6.8	18.2	17.8	18.5
Kuala Lumpur	0.7	0.7	0.6	139	139	7.8	8.4	9.2	9.9	22.9	21.0	20.9
Tanah Rata	0.7	0.6	0.7	49	52	7.3	7.7	10.2	10.8	10.1	19.3	9.4
Johannesburg	0.4	0.4	0.4	101	109	1.9	2.1	4.5	4.6	34.1	20.6	39.4
Kitwe	4.2	3.6	3.5	464	463	26.8	27.3	12.7	12.7	32.8	35.8	36.4
Magoye	1.1	0.4	0.5	25	28	1.9	2.0	4.7	5.4	8.4	8.8	7.8
Harare	0.5	0.4	0.6	194	192	3.4	3.5	4.2	3.9	8.8	8.1	8.1

sented in a paper within this special volume of Water, Air and Soil Pollution dedicated to the Acid Rain 2005 conference (Kucera et al., 2005). However, due to the relatively high measured SO<sub>2</sub> levels it is more correct to use functions for the SO<sub>2</sub>-dominating situation developed within the International co-operative programme on effects on materials including historic and cultural monuments (ICP Materials). Functions from this program are available for zinc, copper and limestone but not for carbon steel (Tidblad et al., 2001),

$$\begin{aligned} \text{ML}_{\text{Zn}} &= 1.4[\text{SO}_2]^{0.22} e^{0.018\text{Rh} - 0.021(T-10)} + 0.029\text{Rain}[\text{H}^+] & T > 10^\circ\text{C} \\ \text{ML}_{\text{Cu}} &= 0.0027[\text{SO}_2]^{0.32} [\text{O}_3]^{0.79} \text{Rh} \cdot e^{-0.032(T-10)} 0.050\text{Rain}[\text{H}^+] & T > 10^\circ\text{C} \\ R_{\text{Limestone}} &= 2.7[\text{SO}_2]^{0.48} e^{-0.018T} + 0.019\text{Rain}[\text{H}^+] \end{aligned}$$

where ML is the mass loss after 1 year of exposure in g/m<sup>2</sup>, *R* is the surface recession after 1 year of exposure in μm, [SO<sub>2</sub>] is the SO<sub>2</sub> concentration in μg/m<sup>3</sup>, [O<sub>3</sub>] is the O<sub>3</sub> concentration in μg/m<sup>3</sup>, Rh is the relative humidity in %, *T* is the temperature in °C, Rain is the amount of precipitation in mm/year and [H<sup>+</sup>] is the hydrogen ion concentration in precipitation in mg/l. For carbon steel a function developed within ISO TC 156 *Corrosion of metals and alloys* has been used (Mikhailov, Tidblad, & Kucera, 2004)

$$\begin{aligned} R_{\text{Steel}} &= 1.77(0.8[\text{SO}_2])^{0.52} e^{0.020\text{Rh} - 0.054(T-10)} \\ &\quad + 0.102D_{\text{Cl}}^{0.62} e^{0.033\text{Rh} + 0.040T} \\ &T > 10^\circ\text{C} \end{aligned}$$

where *D*<sub>Cl</sub> is the chloride deposition in mg m<sup>-2</sup> day<sup>-1</sup>. The results of applying these functions are shown in Table 5 together with the observed values. The values are given in a common unit (μm) for all materials by

**Table 4** Rank correlations between environmental (Table 2) and corrosion (Table 3) data

	SO <sub>4</sub> <sup>2-</sup>	SO <sub>2</sub>	HNO <sub>3</sub>	-pH
Paint coated steel	0.55	–	–	–
Carbon steel	0.85	0.74	–	–
Zinc <sup>a</sup>	0.52	–	–	0.68
Copper	0.62	–	–	–
Limestone <sup>b</sup>	0.62	0.58	0.60	0.80

<sup>a</sup> Kitwe excluded; <sup>b</sup> Johannesburg excluded

**Table 5** Lower and upper quartiles of observed and predicted values using RAPIDC environmental data and dose–response functions (see text) from other exposure programmes

	Observed	Predicted	R <sup>2</sup>
Carbon steel	17–28 μm	9–22 μm	0.79
Zinc <sup>a</sup>	0.5–1.0 μm	0.7–1.3 μm	0.58
Copper	0.7–1.5 μm	0.5–1.1 μm	0.27
Limestone <sup>b</sup>	9–20 μm	3–10 μm	0.74

<sup>a</sup> Kitwe excluded; <sup>b</sup> Johannesburg excluded

using the density of carbon steel (7.8 g cm<sup>-3</sup>), zinc (7.14 g cm<sup>-3</sup>) and copper (8.93 g cm<sup>-3</sup>). For carbon steel, copper and limestone the predicted values are lower but for zinc the observed values are lower. One explanation of the low zinc values can be that this material is particularly sensitive to the starting conditions of the exposure. It is worth noting the substantial difference between observed and predicted values for Portland limestone. For copper the comparison is misleading since the 1-year values cannot be predicted by the proposed equation (*R*<sup>2</sup>=0.27). Therefore, the 2- and 4-year results will be very valuable in the assessment of corrosion rates in the RAPIDC network compared to results from other networks in Europe.

## 4 Conclusions

A network of organisations and corrosion test sites have been formed with six partners (12 sites) in Asia and three partners (4 sites) in Africa and an exposure programme was started in May–August 2002. Complete results of an extensive environmental characterisation and corrosion data of painted steel, carbon steel, zinc, copper and limestone after one year of exposure in the network have been presented and the conclusions below are preliminary in anticipation of results after 2 and 4 years of exposure. SO<sub>2</sub> pollution is the most decisive factor but in addition a correlation

to pH has been found for zinc and limestone and a correlation to HNO<sub>3</sub> has been found for limestone. Based on best available dose–response functions the corrosion values are higher than expected except for zinc where the values are lower than expected. For limestone the values are substantially higher than expected. For painted steel and copper 1 year of exposure is too short and a comparison can be misleading. Therefore, future studies will include a complete evaluation of the 2- and 4-year results that will be very valuable in the assessment of corrosion rates in the RAPIDC network compared to results from other networks in Europe.

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