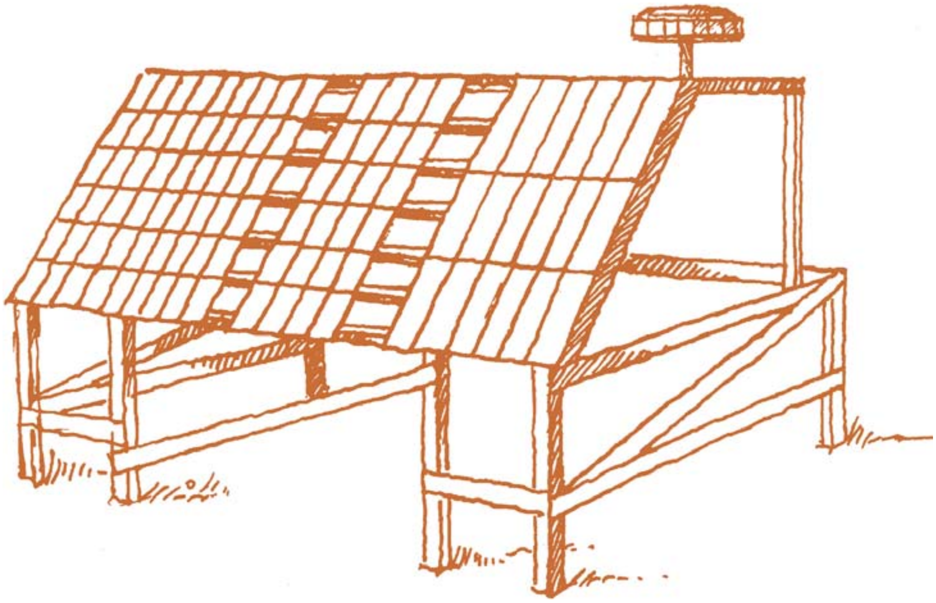
CONVENTION ON LONG-RANGE TRANSBOUNDARY AIR POLLUTION

UN/ECE INTERNATIONAL CO-OPERATIVE PROGRAMME ON EFFECTS ON MATERIALS, INCLUDING HISTORIC AND CULTURAL MONUMENTS



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Results from the multipollutant programme: Corrosion attack on painted steel after 1, 2 and 4 years of exposure (1997-2001)

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Results from the multipollutant programme: Corrosion attack on painted steel after 1, 2 and 4 years of exposure (1997-2001)

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Summary

International Co-operative Programme on Effects on Materials, including Historic and Cultural Monuments is a research project launched by the Executive Body for the Convention on Long-range Transboundary Air Pollution for studying the effect of airborne pollution on materials. The study is in its second phase and this report covers the results after one, two and four years of exposure of painted panels.

The paint system exposed has been steel panels coated with two layers alkyd paint (total 80 µm).

The damage of the paint system has been evaluated by using the well-established ASTM-standards on samples exposed.

Damage from the cut is observed at all test sites as early as after one year. After four years ASTM values from 8 to 5 are observed. The highest damage of painted panels is observed on sites with high air pollution like site 3 Kopisty and on rural sites with high amount of rain like site 23 Birkenes. The high damage observed on site 44, Svanvik, illustrates the effect of short high level of SO_2 in an area where the monthly average still is low and the washing off by rain is moderate. As an illustration, the average concentration for the winter period 2001-2002 was 5.5 µg/m^3 but during that time it was observed 9 days with daily average higher than 50 µg/m^3 and 53 hourly values higher than 100 µg/m^3.

Filiform corrosion, shown as thin whiskers under the paint film from the cut, are more dominating in the second phase of the programme. They turn up on all the new sites in the programme, but only for 1/2 of the sites from the first phase. High humidity is described in the literature as an important factor for filiform corrosion and some of the results may support this observation, but the multipollution situation observed in big cities, may also be important. For the comparison between the two different exposure periods there are two factors that could complicate the comparison. Another company has painted the samples. The second concern is the paint itself. The paint should be the same, but with 8 year's difference in the paint production, even the same formula could have some variation during this period.

At several places the paint surface was cracked after four years. For some of the sites it could be seen without a microscope. The worst cracked paint was observed on site 43 Tel Aviv, where the cracks went down to the steel substrate and the paint was spotted with corrosion products. The effect is more dominating on sites with high temperature in the Mediterranean area than in Northern Europe.

A comparison between the results from the first and second phase shows that the damages are less severe this time. The same trend has earlier been shown for the air pollution parameters. The reduction is highest in the most polluted sites like Milan and Kopisty but it is also substantial at the rural EMEP sites Birkenes, Aspvreten and Ähtäri in Scandinavia. It is therefore most likely that the reduced level of air pollution causes the reduced corrosion of painted panels. The dose-response

equations reported at the end of phase one tried to define the lifetime before maintenance was needed using ASTM ≤ 5. To run a new statistical evaluation for dose-response correlation more data is needed. The interaction with the filiform deterioration also complicates the evaluation and the evaluation has therefore been dropped.

Results from the multipollutant programme: Corrosion attack on painted steel after 1, 2 and 4 years of exposure (1997-2001)

1. Introduction

Airborne acidifying pollutants are known to be one major cause of corrosion of different materials including the extensive damage that has been observed on historic and cultural monuments. In order to fill some important gaps of knowledge in this field the Executive Body for the Convention on Long-range Transboundary Air Pollution decided to launch an international co-operative programme. The programme was started in September 1987 and has involved exposure at 39 test sites in 12 European countries and in the United States and Canada. The first phase of the exposure programme was finished in 1995. However, during the eight years where the exposure programme has been carried out, a large change in the pollution situation in Europe has been observed. The SO_2 concentrations have been drastically reduced, while the changes in the NO_2 and O_3 levels have been minor. This new pollution situation where the importance of NO_2 and O_3 were in focus, led to a proposal of a second phase of the programme. The new 4 years exposure project was launched in the fall 1997 with redefined environmental measuring programme, a better combination of test sites for field exposure and with several new countries as partners in the project (Swedish Corrosion Institute rev. 1993).

The aim of the new programme is to perform a quantitative evaluation of the effect of NO_x and other pollutants like ozone and sulphur pollutants in combination with climatic parameters on the atmospheric corrosion of important materials. For this purpose, measurements of gaseous pollutants, precipitation and climate parameters have been initiated at or nearby each test site, together with evaluation of corrosion of the exposed test materials at each site.

A Task Force is organising the programme with Sweden as lead country and Swedish Corrosion Institute serving as the Main Research Centre. Sub-centres in different countries have been appointed, each responsible for their own material group. The material groups are:

Structural metals:

- Steel and zinc for trend analyses (Sub-centre responsible for evaluation: SVUOM Praha as., Prague, Czech Republic).
- Zinc for 4 years of exposure (EMPA Corrosion/Surface Protection, Dübendorf, Switzerland).
- Copper and cast bronze (Bayerisches Landesamt für Denkmalpflege, Munich, Germany).

Stone materials: Portland limestone (Building Research Establishment Ltd., Department of Environment, Waterford, United Kingdom).

Paint coatings: Steel with silicon alkyd paint (Norwegian Institute for Air Research, Kjeller, Norway).

Glass materials: Two types of glass M1 and M3 (Institute of Chemistry, Academy of Fine Arts, Vienna, Austria)

Norwegian Institute for Air Research has been the sub-centre for the environmental database through the whole programme.

The exposure programme has fewer materials than in the first phase, mainly because we have to use materials that are sensitive enough for having sufficient reaction within 4 years of exposure.

2. Materials

The paint system tested was steel panels coated with two layers of alkyd paint (total 80 µm). The edges were sealed with an extra thick coating to prevent corrosion from the edges.

3. Evaluation

The test panels have a horizontal cut on the front surface and are painted with alkyd on both sides. The painted panels are only freely exposed since the expected lifetime in sheltered position is much longer than 4 years.

The evaluation has been focused on the spread of damage around an artificial cut through the paint measured in mm and given as the ASTM rating numbers from RN = 10 to RN = 0 (ASTM 1987). Beside the ordinary spread from the cut, filiform corrosion is observed around the cut on several of the test panels. Evaluation techniques for filiform corrosion are described in ISO standard 4623 (ISO 2000). The evaluation techniques have been different for the two corrosion types and this is illustrated in Figure 1.

The results of evaluation are presented in different way for the two types of attack. For spread from cut the total spread (a_1) excluding the filiform part is measured. The results are given as the mean spread of the three samples (a_m) in one direction after subtracting the width of the cut (1 mm)

$$a_m=[(a_1+ a_2+ a_3)-3]/6$$

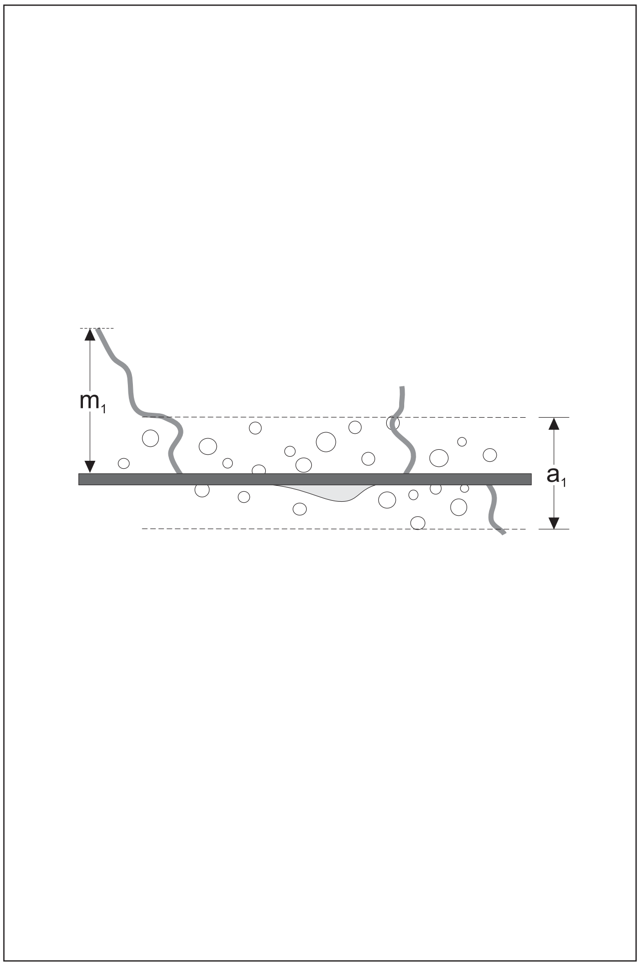

Figure 1: Illustration of evaluation for spread from the cut and filiform corrosion on the test panels.

The results of the evaluation in mm and the corresponding ASTM value for one, two and four years are given in Table 1.

For filiform corrosion, the longest tread (m_1) for each test sample is measured. The mean value (m_m) in mm for the three parallels is given in Table 2. For sites where only one of the panels has filiform corrosion this results are specially marked.

$m_m =(m_1 + m_2 + m_3)/3$

Other forms of damages are only reported when the damages seem important for the total evaluation of the attack.

4. Results

4.1 Damage located to the cut

Table 1 gives the results for 1, 2 and 4 years of exposure. Samples 35B and 40B are exposed with the cut facing down.

Table 1: The results for spread from cut for the alkyd painted panels after one, two and four years of exposure.

			1997-98	1997-98	1997-99	1997-99	1997-01	1997-01
Site no	Country	Place	mm	ASTM D 1654-79a	mm	ASTM D 1654-79a	mm	ASTM D 1654-79a
01	Czech Republic	Prague- Bech.	1.2	7	1.6	7	2.8	6
03	Czech Republic	Kopisty	2.3	6	2.5	6	3.5	5
05	Finland	Äthäri	0.8	8	1.7	7	1.8	7
07	Germany	Waldhof L.	1.7	7	2.6	6	2.7	6
09	Germany	Langenfeld	1.3	7	1.3	7	2.4	6
10	Germany	Bottrop	1.4	7	1.9	7	3.6	5
13	Italy	Rome	0.8	8	0,8	8	*	*
14	Italy	Casaccia	1.2	7	1.3	7	0.9	8
15	Italy	Milan	1.4	7	1.3	7	1.8	7
16	Italy	Venice	1.1	7	1.3	7	1.6	7
21	Norway	Oslo	0.7	8	1.3	7	1.2	7
23	Norway	Birkenes	2.3	6	2.9	6	3.6	5
24	Sweden	Stockholm	1.3	7	1.4	7	1.7	7
26	Sweden	Aspvreten	2.3	6	2.3	6	2.7	6
27	United Kingdom	Lincoln Cathedral	3.0	6	2.8	6	2.6	6
31	Spain	Madrid	0.3	9	0.6	8	1.2	7
33	Spain	Toledo	0.4	9	0.7	8	1.4	7
34	Russia	Moscow	1.5	7	1.4	7	1.4	7
35	Estonia	Lahemaa	2.6	6	4.0	5	2.3	6
35 B	Estonia	Lahemaa (cut down)	1.2	7	1.8	7		
36	Portugal	Lisbon	2.8	6	3.4	5	2.4	6
37	Canada	Dorset	1.3	7	2.2	6	2.6	6
40	France	Paris	1.8	7	1.9	7	0.9	8
40 B	France	Paris (cut down)	0.7	9	1.1	7		
41	Germany	Berlin	1.7	7	1.3	7	1.5	7
43	Israel	Tel Aviv	1.6	7	1.9	7	1.7	7
44	Norway	Svanvik	1.6	7	1.7	7	3.8	5
45	Switzerland	Chaumont	2.1	6	1.8	7	2.5	6
46	United Kingdom	London	5.8	4	4.6	5	2.4	6
47	USA	Los Angeles	1.4	7	1.6	7	1.3	7
49	Belgium	Antwerp	2.4	6	1.6	7	1.0	8

* = missing samples

The spread from the cut in mm is reported as the mean distance between the blisters above and below the cut. The first year it can be difficult to distinguish between filiform and spread from cut since also the spread is localised as small paint blisters. After four years when the paint along the cut is lifted by the corrosion products the blisters are less dominating. For some of the test sites this has given less spread from the cut with time even if the general visual impression clearly shows that the paint system has become weaker.

It seems to be a trend to more damage on sites with high pollutant levels and to the sites with more humid conditions.

4.2 Filiform corrosion

While flaking and blistering were the dominating form for damage in the first phase of the programme, filiform attack occurs more frequently in the second phase. Figure 2 shows a typical example of this type of attack taken from the two years of exposure at Lincoln Cathedral. Table 2 gives the results of filiform corrosion for the test sites and the evaluation technique used is illustrated in Figure 1.

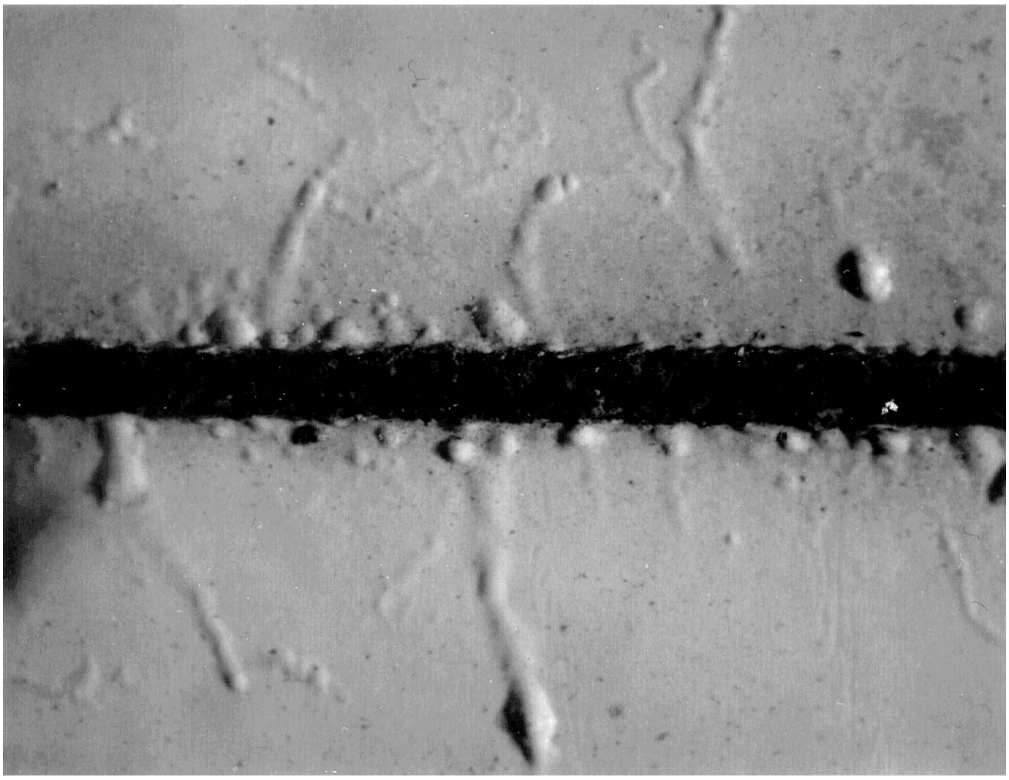

Figure 2: Filiform attack from the cut on alkyd painted panels at site Lincoln Cathedral.

In the evaluation the results can be divided into two groups. First group is panels without any sign of filiform corrosion marked “none” in Table 2. Second group is panels where threads are observed. The spread from the cut of the longest thread is measured in millimetre in the same way as the measurement of m_1 in Figure 1.

Filiform corrosion may some time be hard to detect by the eye. In our inspection microscope and light from the side has been used, this makes it much easier to detect the filiform corrosion.

Filiform attack will reach further out from the cut than the blisters and the number of sites with filiform corrosion is increasing with the exposure time as seen in Table 2. Filiform corrosion is complicated to evaluate since it is large deviation been the parallels. Further the distinctness of the filiform corrosion differed from one panel to the next. The filiform threads observed develop in different directions from the cut. When the measurements are made perpendicularly to the cut the variation between the results from the parallels will be higher. According to the ISO-standard the results are often described in more general terms like "slight", "moderate" and "severe". Often filiform corrosion may occur only on one of the three panels. This is marked in Table 2

Obvious the filiform corrosion is a corrosion type related to the cut and could therefore be a part of damage from cut result. However the established ASTM scale has no relation to the filiform corrosion and therefore the results are presented separately.

Filiform corrosion was almost never observed in the first phase of the ICP materials programme, while it was observed after one year in the second phase. It was a remarkable difference in the results between the sites that were included also in the first phase ("old sites") and the new sites only present in the second phase ("new sites"). All the new sites had filiform corrosion after one year while only 7 of 21 among the old sites had filiform corrosion on all three panels. Even after four years 8 of the old sites had no filiform corrosion. Site 40 Paris has exposed panels both with the cut up and down. The filiform corrosion was strong on the panels facing down, but weak on panels facing up. The four years panels from site 40 have been partly exposed in both directions.

Table 2: The results for filiform attack on the alkyd painted panels after one, two and four years of exposure.

			1997-98	1997-99	1997-01
Site no	Country	Place	mm	mm	mm
01	Czech Republic	Prague- Bech.	none	none	none
03	Czech Republic	Kopisty	none	none	none
05	Finland	Ähtäri	2 (only one panel)	none	none
07	Germany	Waldhof L.	none	4.3	4.8
09	Germany	Langenfeld	2.7	3.3	3.3
10	Germany	Bottrop	3.0 (only one panel)	3.0	2.8
13	Italy	Rome	none	none	*
14	Italy	Casaccia	5.8	6.7	4.7
15	Italy	Milan	none	none	none
16	Italy	Venice	3.0 (only one panel)	5.5	4.3
21	Norway	Oslo	none	none	none
23	Norway	Birkenes	9 (only one panel)	none	6.5 (only one panel)
24	Sweden	Stockholm	none	none	none
26	Sweden	Aspvreten	6.0 (only one panel)	3.0	2.5
27	United Kingdom	Lincoln Cathedral	10.2	11.5	13.2
31	Spain	Madrid	none	none	none
33	Spain	Toledo	none	none	none
34	Russia	Moscow	7 (only one panel)	3.0 (only one panel)	2.5
35	Estonia	Lahemaa	1.8	none	2.7
35 B	Estonia	Lahemaa (cut down)	3.0	2.0	
36	Portugal	Lisbon	10.5	7.5	8.7
37	Canada	Dorset	2.0	3.0	3.8
40	France	Paris	none	6.0 (only one panel)	8.0
40 B	France	Paris (cut down)	7.5	8.5	
41	Germany	Berlin	7.7	8.8	4.8
43	Israel	Tel Aviv	9.3	5.0	6.3
44	Norway	Svanvik	6.7	7 (only one panel)	9.2
45	Switzerland	Chaumont	5.5 (only one panel)	2.0	5.5
46	United Kingdom	London	11	14.3	15.7
47	USA	Los Angeles	7.2	3.8	3.5
49	Belgium	Antwerp	8.5	9.3	7.5

4.3 General appearance

A visual inspection of the paint condition after the four years of exposure gave the following impression. Black surface fungus gave the most dominating bad impression. At some sites the fungus was mixed with algae. Only on site 3 Kopisty soot was observed together with the fungus. It was also interesting to see that the visual impression was different on the back and front side, and the rating between the sites also became different. The best appearance was observed on sites where chalking gave a nice white impression of the paint surface. Ranking of the worst and the best appearance is given in Table 3.

Table 3: Ranking of the general appearance for the front and backside.

Ranking	Site nr-Front side	Site nr-Back side
Worst	49 Fungus	27 Fungus
↓	16 Fungus	49 Fungus
	43 Rusty surface	41 Fungus
	23 Fungus	23 Fungus
	3 Soot and Fungus	7 Fungus
	27 Fungus	36 Fungus
	14 Fungus	16 Fungus
	41 Fungus	10 Fungus
Best	33,24,40,44,46 chalking	33,40,44

4.4 Cracking

At several places the paint surface was cracked. For some of the sites it could be seen without a microscope. The worst cracked paint was observed on site 43 Tel Aviv. At that site the cracking went down to the steel substrate and the paint was spotted with rust coming through the paint film, see Figure 3-Figure 5.

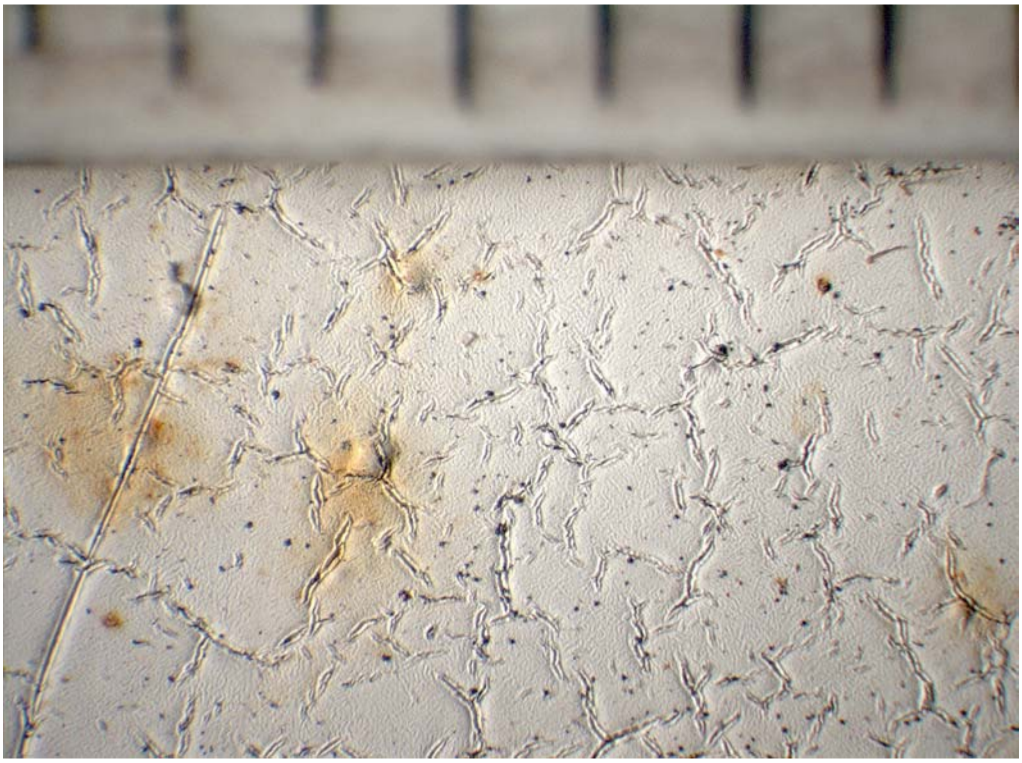

Figure 3: The paint surface after four years of exposure at site 43 Tel Aviv.

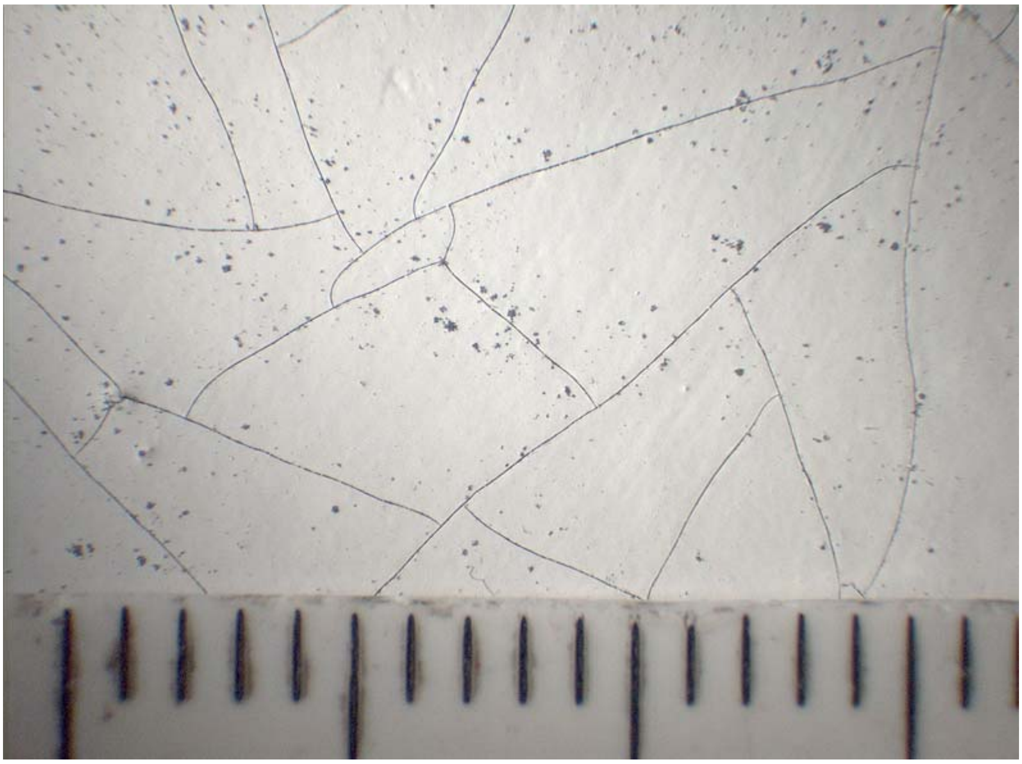

Figure 4: The paint substrate after four years of exposure at site 37 Dorset, Canada.

Figure 5: The paint substrate with cracks after four years of exposure at site 16 Venice.

5. Trends

In Table 4 the results of damage from cut from the test sites that are included in both phases of the programme, are given. The results listed are the values from one, two and four years of exposure. The table can be used for comparison of trends for the different sites. The results from the first phase of the programme (1987-88) are earlier reported (Henriksen et al., 1998).

In Figure 6 a scatter plot is showing the results after four years of exposure for the first and second phase of the programme.

Table 4: The results from the alkyd painted panels after one and two years of exposure in 1997-99 and 1987-89.

			1997-98	1987-88	1997-99	1987-89	1997-2001	1987-91
Site no	Country	Place	mm	mm	mm	mm	mm	mm
01	Czech Republic	Prague-Bech.	1.2	2.7	1.6	4.8 (x)	2.8	4.7
03	Czech Republic	Kopisty	2.3	6.2	2.5	8.3	3.5	17.8
05	Finland	Ähtäri	0.8	2.3	1.7	2.9	1.8	3.2
07	Germany	Waldhof L.	1.7	2.9	2.6	3.3	2.7	4.4
09	Germany	Langenfeld	1.3	1.8	1.3	2.4	2.4	3.1
10	Germany	Bottrop	1.4	3	1.9	3.3	3.6	4 (x)
13	Italy	Rome	0.8	0.8	0.8	1.1	missing	1
14	Italy	Casaccia	1.2	1.8	1.3 (x)	2.1	0.9	3.2 (x)
15	Italy	Milan	1.4	4.2	1.3	5.5	1.8	8.3
16	Italy	Venice	1.1	2.2	1.3	2.9	1.6	5.2
21	Norway	Oslo	0.7	1.9	1.3	2.3	1.2	2.8
23	Norway	Birkenes	2.3	3.8	2.9	5.3	3.6	6.3
24	Sweden	Stockholm	1.3	2.1	1.4	2.2	1.7	2.4
26	Sweden	Aspvreten	2.3	2.6	2.3	3.4	2.7	3.8
27	United Kingdom	Lincoln Cathedral	3.0	2.5	2.8	2.3	2.6	2.4
31	Spain	Madrid	0.3	1.9	0.6	2	1.2 (x)	2.7
33	Spain	Toledo	0.4	0.8	0.7	0.8	1.4	1.1
34	Russia	Moscow	1.5	1.0	1.4	1.2	1.4	1.8
35	Estonia	Lahemaa	2.6	3.3 (x)	4.0	2.9	2.3	3.7
36	Portugal	Lisbon	2.8	1.7	3.4	1.6	2.4	1.3
37	Canada	Dorset	1.3	1.7	2.2	2.5	2.6 (x)	3.4

(x) = large spread between the parallels

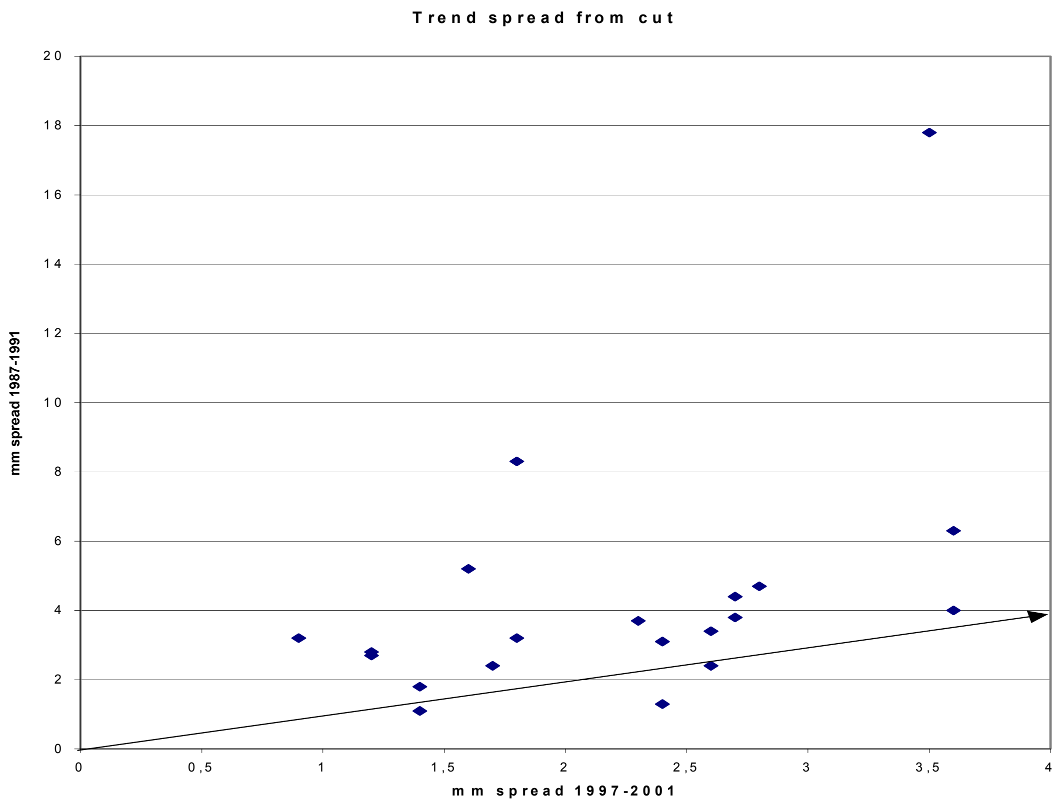

Figure 6: The spread in damage from cut after four years of exposure for the period 1987-91 against 1997-2001.

From the data in Table 4 and Figure 6 it is obvious that most sites have had a positive trend for the last ten years. A slight change to more corrosion is observed on the sites 27 Lincoln Cathedral, site 33 Toledo and site 36 Lisbon. Particularly the sites with high attack in the first phase have got the largest reduction.

6. Discussion

An evaluation of the deterioration of paint by use of ASTM standards has some limitations. Even if the deterioration processes themselves are irreversible, the evaluation techniques based on visual inspections of changes around a cut using simple tools are not able to record the processes correctly. A major problem is that the processes interact with each other and the environment can affect the resulting damages in different ways in different climates. An example is the Tel Aviv site where the cracking of the paint occurs in a way never observed in the first phase of the programme.

Filiform corrosion, shown as thin whiskers under the paint film from the cut, are more dominating in the second phase of the programme. They turn up on all the new sites in the programme, but only for 1/2 of the sites from the first phase. High humidity is described in the literature as an important factor for filiform corrosion and some of the results may support this observation, but the multipollution situation observed in big cities, may also be important. For the comparison between the two different exposure periods there are two factors that could complicate the comparison. Another company has painted the samples. The second concern is the paint itself. The paint should be the same, but with 10 years difference in the paint production, even the same formula could have some variation during this period.

For the statistical dose-response evaluation we have chosen the deterioration parameter damage from the cut since this was the parameter where the results were expected to develop first. It is also an irreversible process that is easy to follow. Damage from the cut is observed at all test sites and after as early as after one year After four years ASTM values from 8 to 5 are observed. The highest damage of painted panels is observed on sites with high air pollution like site 3 Kopisty and on rural sites with high amount of rain like site 23 Birkenes. The high damage observed on site 44 Svanvik illustrates the effect of short high level of SO_2 in an area where the monthly average still is low and the washing off by rain is moderate. As an illustration, the average concentration for the winter period 2001-2002 was 5.5 µg/m^3 but during that time it was observed 9 days with daily average higher than 50 µg/m^3 and 53 hourly values higher than100 µg/m^3. (Hagen et al., 2002).

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A comparison between the results from the first and second phase shows that the damages are less severe this time. The same trend has earlier been shown for the air pollution parameters. The reduction is highest in the most polluted sites like Milan and Kopisty but it is also substantial at the rural EMEP sites Birkenes, Aspvreten and Ähtäri in Scandinavia. It is therefore most likely that the reduced level of air pollution causes the reduced corrosion of painted panels. The dose-response equations reported at the end of phase one tried to define the lifetime before maintenance was needed using ASTM $\leq$ 5 (Henriksen et al., 1998). To run a new statistical evaluation for dose-response correlation more data is needed. The interaction with the filiform deterioration also complicates the evaluation and the evaluation has therefore been dropped.

7. Conclusions

The damage of the paint system has been evaluated by using the well-established ASTM-standards on the samples exposed. The results show that the damage from the cut is less dominating in the second phase of the programme. This follows the trends also observed for the pollution parameters.

Filiform corrosion is more domination around the cut in the second phase and particularly among the new sites in phase two. The paint system should be the same in both phases. However, there is ten year’s difference between the paint productions and the application is made by another company this time. These differences will increase the uncertainty in the evaluation.

The development of the damages is still too low for estimating a lifetime for the paint system in different environments and to estimate a dose-response equation for the environmental impact.

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ABSTRACT

The paint steel panels coated with two layers alkyd paint (total 80 µm) have been exposed for 1, 2 and 4 years. The damage of the paint system has been evaluated by using the well-established ASTM-standards.
Damage from the cut is observed at all test sites, and after four years ASTM values from 8 to 5 are observed. Comparing the results with the environmental parameters seems to indicate that the damage is affected by air pollution like in the big cities. However, the damage is also high at rural sites, particularly at sites with high amount of rain.

Filiform corrosion, shown as thin whiskers under the paint film from the cut, is more dominated in the second phase of the programme than in first phase 1987-1991. Cracking of the corrosion protection paint films has been observed on several samples after four years particularly among the Mediterranean sites.

NORWEGIAN TITLE

Korrosjon på malt stål etter 1, 2 og 4 års eksponering.

KEYWORDS		
Paint	Damage	Air pollution

ABSTRACT (in Norwegian)

* *Classification*   *A*   *Unclassified (can be ordered from NILU)*
B   *Restricted distribution*
C   *Classified (not to be distributed)*