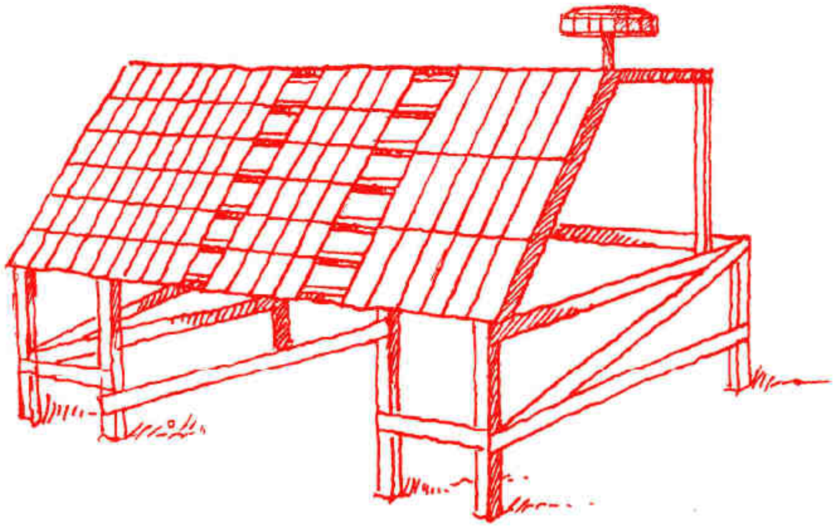
CONVENTION ON LONG-RANGE TRANSBOUNDARY AIR POLLUTION

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



Report No. 47

RESULTS FROM THE MULTIPOLLUTANT PROGRAMME:
TREND OF CORROSIVITY BASED ON CORROSION RATES (1987-2001)

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Summary

This report summarizes the results of evaluation of weight changes and corrosion losses of carbon steel and zinc after 1 year of exposure from 30 test sites in period 1997/98 and 2000/01 in frame of UN ECE ICP Effects on Materials. Results are compared with previous 1 year´s exposures in periods 1987/88, 1992/93 and 1994/95 to evaluate trends in corrosivity of atmospheres connected with decreasing of pollution of atmospheres by SO_2.

The results for corrosion loss for the last two exposure period for carbon steel and zinc are presented in Table 3-6. The comparison of all exposure periods is presented in Table 10-11.

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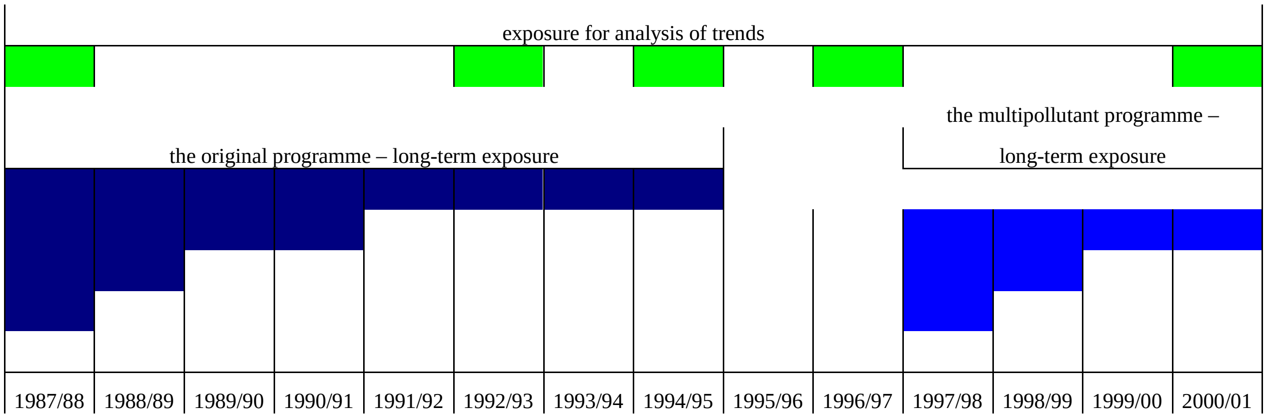
Introduction

The UN ECE International Co-operative Programme was performed to a quantitative evaluation of the effect of sulphur pollutants in combination with NO_x and other pollutants as well as climatic parameters on the atmospheric corrosion of important materials. The programme was based on an international field exposure on 39 test-sites in a wide geographical zone of the 13 Signatories of the Convention on Long-Range Transboundary Air Pollution. The basic field exposure was performed in 1987-1995.

Within 1985 - 1995 the environmental situation have been changed. The SO_2 concentrations significantly decreased in many European countries. During the Programme Task Force meeting in Lillestrom (May 27-29, 1991) was proposed to use the ICP UN ECE test sites for long-term observation of trends in corrosivity changes by means of a limited number of exposed materials for one-year periods.

The first repeated one-year exposure of steel and zinc samples was realised in autumn 1992 and the second exposure in autumn 1994. In 1997 the multipollutant exposure programme has opened. Only selected test sites from the first stage of programme and new countries with new test sites were included in this programme. The new programme includes 18 countries with 30 test sites. In autumn 1996 and 2000 one-year exposures of unalloyed steel and zinc in open atmosphere were started to follow trends and to estimation of corrosivity of new test sites.

Time schedule of exposure programme



This report introduces the comparative evaluation of the results for periods

- 1987-1988, 1992-1993, 1994-1995, 1996-1997, 1997-98 and 2000-01 for carbon steel samples
- 1989-1990, 1992-1993, 1994-1995, 1996-1997 and 2000-01 for zinc samples.

SVUOM has been subcentre responsible for exposure and evaluation of samples of carbon steel and zinc for determination of trends in corrosivity of atmospheres in the frame of programme.

1. Methods

1.1. Characterisation of samples

Plates of unalloyed carbon steel (with C < 0.2 %, P < 0.07 %, Cr < 0.07 % according to CSN 11373) and zinc (98,5 %) with dimensions 100 x 150 x 0,5 mm were used, triplicate per exposure period.

Before the exposure the samples were degreased in alkaline degreaser, rinsed with ethanol, dried and weighed.

1.2. Exposure of samples

Methods prescribed in the Technical manual to the UN ECE International Co-operative Programme on Effects on Materials including Historic and Cultural Monuments (Report No 1, June 1988) were applied for exposure of samples on test sites. Samples were exposed on racks in open atmosphere (unsheltered exposure) according to ISO 8565.

The list of test sites for the multipollutant programme is in Table 1 and Figure 1. The new test site No 50 Katowice (Poland) was introduced for exposure period 2000/01. Exposure period has been one year. The field exposure evaluated in this report included results from exposure of carbon steel in periods 1996/97, 1997/98 and 2000/01 and of zinc in period 1996/97 and 2000/01.

1.3. Measurement of environmental data

Basic climatic parameters (temperature, relative humidity and intensity of sunshine radiation), concentration of gaseous pollution (SO_2, NO_x, O_3) and precipitation (amount, pH, conductivity, amount of SO_4^{2-}, NO_3^-, Cl^-, and NH_4^+, Na^+, K^+, Ca^{2+}, Mg^{2+} if available) were measured on each test site. All data were reported to, and completed by the Norwegian Institute for Air Research - NILU, which also checked the quality of the data.

1.4. Evaluation of corrosion attack

The visual evaluation of layer of corrosion products was done to indicate non uniform corrosion process.

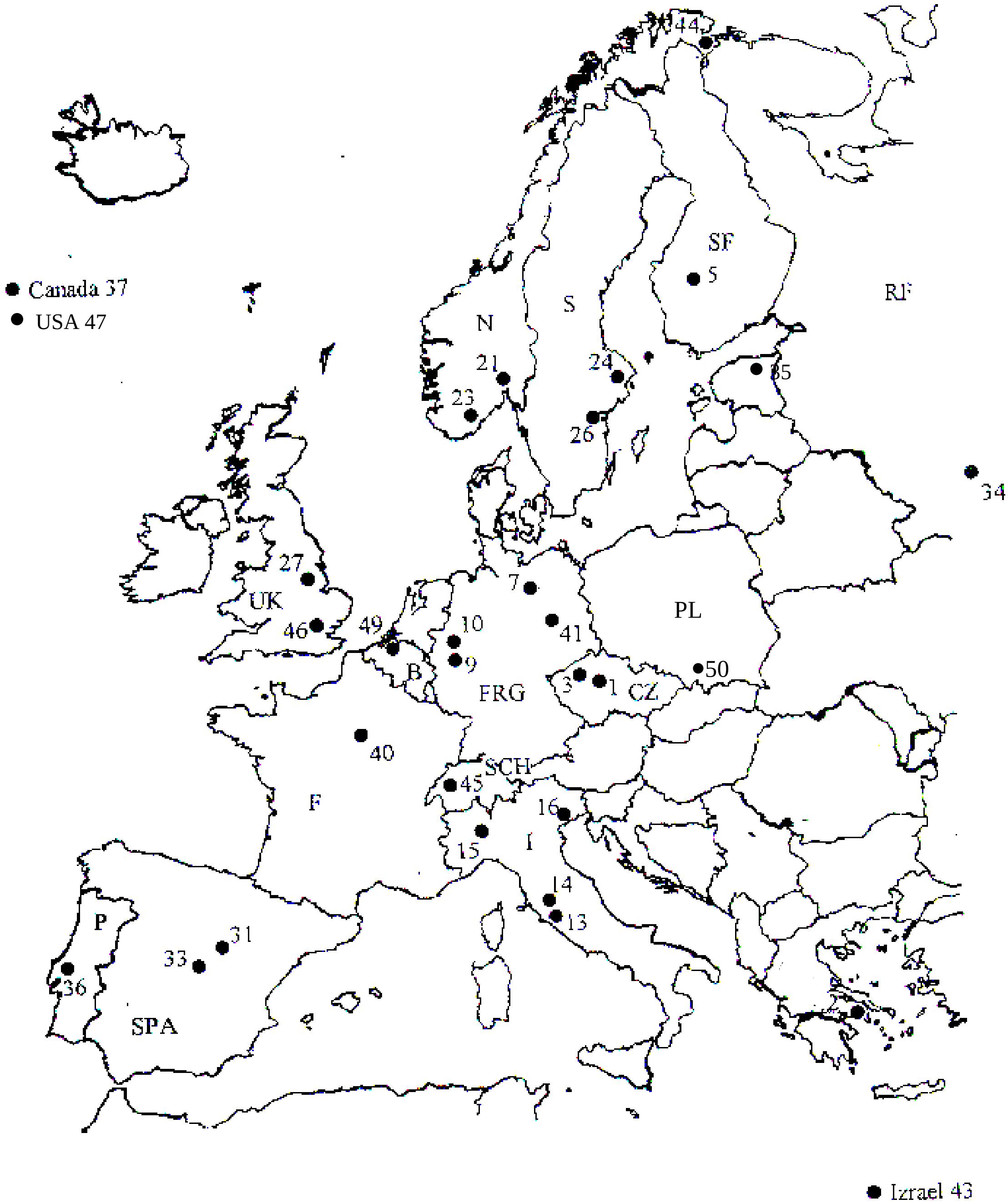
After exposure samples were weighed and the corrosion products were removed using pickling method according to ISO 8407 (Annex A - solution C3.5. and C9.5.). The corrosion losses had been obtained by gravimetric method.

For selection samples of carbon steel and zinc analysis of corrosion products was performed by spectrophotometric analysis of water extract of layer of corrosion products. There were analysed corrosion products from 1 sample exposed at test sites No 3, 10, 13, 40, 41, 43, 46 and 50, it means test sites with the highest corrosion losses.

Table 1 - The list of test sites

Country	Test site	Type of atmosphere
the Czech Republic	1 Prague 3 Kopisty	urban-industrial industrial
Finland	5 Ahtari	rural
the Federal Republic of Germany	7 Waldhof-Langenbrugge 9 Langenfeld 10 Bottrop 41 Berlin	rural rural industrial urban
Italy	13 Rome 14 Casaccia 15 Milan 16 Venice	urban rural urban-industrial urban
Norway	21 Oslo 23 Birkenes 44 Svanvik	urban rural rural
Sweden	24 Stockholm South 26 Aspvreten	urban rural
the United Kingdom	27 Lincoln Cathedral 46 London	urban urban
Spain	31 Madrid 33 Toledo	urban rural
the Russian Federation	34 Moskow	urban-industrial
Estonia	35 Lahemaa	rural
Portugal	36 Lisbon	urban
Canada	37 Dorset	rural
France	40 Paris	urban
Israel	43 Tel Aviv	urban
Switzerland	45 Chaumont	rural
USA	47 Los Angeles	urban-marine
Belgium	49 Antverps	urban-marine
Poland	50 Katowice	industrial

Figure 1 – Map showing location of test sites



1.5. Runoff

The runoff of zinc had been evaluated for samples exposed in period 2000/01 by gravimetric method [1, 2].

2. Results

2.1. Visual evaluation

The layers of corrosion products of carbon steel and zinc had formed a typical structure, colour and thickness according exposure conditions in period 1996/97. The surface layer of samples of carbon steel exposed on test site No. 43 had been affected by erosion (sand storm).

On carbon steel and zinc samples exposed in period 2000/01 at test site No 23 there were found a specific corrosion products formed in wet conditions without air circulation. The contact person informed that the samples were withdrawn as frozen and packed. The surfaces of all zinc samples exposed in period 2000/01 at the test site No 36 were contaminated by non uniform white layer which is not typical corrosion product of zinc.

The basic information about corrosion behaviour of carbon steel and zinc in different environmental conditions gives the value of weight change and mass loss.

2.2. Weight changes of carbon steel and zinc

Weight change of exposed samples had been started to evaluate from exposure period 1996/97. The measured value represents a differences of mass of corrosion products originated at impact of environment and part of them removed by run-off erosion and fall out of non adherent particles and sub-layers. Mass increase of weight of exposed steel samples gives a complex and not well-defined information. Weight changes of exposed samples of carbon steel and zinc are summarised in Table 2 and Figures 2 and 3. The values of weight changes are included also for exposure period 1997/98 in this report.

After exposure in period 1996/97 the mass of unsheltered samples of carbon steel increased during exposure on all test sites except test site No. 10 Bottrop. The mass of unsheltered samples of carbon steel increased during exposure on all test sites except test site No. 43 Tel Aviv for exposure period 1997/98. In period 2000/01 the mass of carbon steel samples increased during exposure with exception of test site No. 27 Lincoln Cathedral.

It is obvious the mass of exposed carbon steel had been on the same level during three exposure periods for the majority of test sites with exceptions:

- the significant decreasing of mass for period 1997/98 and 2000/01 in comparison with the period 1996/97 for test sites No. 1, 31 and 33,
- the mass of samples increased in period 2000/01 in comparison with previous two exposure periods for test site No. 16,
- the significant change had been found between the mass of samples after exposure in period 1996/97 and other two periods for test site No. 41.

After exposure in period 1996/97 the weight changes of unsheltered samples of zinc were negative for all test sites. In period 2000/01 the mass of exposed samples were positive on test sites No. 5, 13, 16, 26, 36, 41 43 and 44. The mass of exposed zinc had been on the same level during two exposure periods for test sites No. 7, 10, 15, 21, 34, 37 and 45. The significant changes was found for test sites No. 3 and 31. During one year exposure the forming of solid corrosion product layer on zinc is not stabilized.

Table 2 – Weight changes (g.m^{-2}) of unalloyed steel and zinc after one year´s exposure in open atmosphere (period 1996/1997, 1997/98 and 2000/01)

Test site	carbon steel			zinc	
	1996/97	1997/98*	2000/01	1996/97	2000/01
1 Prague	77,0	55,7	55,5	- 1,73	- 0,9
3 Kopisty	90,7	79,7	105,0	- 3,17	- 0,8
5 Ahtari	21,7	23,3	21,3	- 0,50	0,6
7 Waldhof-Langenbrugge	36,0	40,0	36,3	- 1,20	- 1,0
9 Langenfeld	18,0	23,0	37,8	-	- 0,4
10 Bottrop	- 2,3	18,7	27,0	- 3,13	- 2,7
13 Rome	40,0	30,0	39,5	- 0,90	0,2
14 Casaccia	39,7	47,0	50,5	- 0,17	0,0
15 Milan	45,0	36,7	34,2	- 1,20	- 1,6
16 Venice	8,3	6,3	23,0	- 0,27	0,5
21 Oslo	37,0	38,7	33,2	- 0,80	- 0,8
23 Birkenes	37,0	33,3	31,7	- 1,50	- 1,1
24 Stockholm South	29,0	38,3	27,2	- 1,10	- 0,5
26 Aspvreten	27,7	-	25,4	- 0,57	0,1
27 Lincoln Cathedral	-	9,0	- 0,4	-	- 1,1
31 Madrid	52,3	33,7	37,8	- 3,43	-0,1
33 Toledo	32,0	26,0	19,3	- 1,40	-0,2
34 Moskow	46,3	34,0	33,6	- 0,57	- 0,9
35 Lahemaa	-	31,7	38,2	-	- 0,5
36 Lisbon	1,3	6,0	4,9	- 0,07	1,0
37 Dorset	50,3	51,7	45,9	- 1,17	- 1,1
40 Paris	-	28,0	24,4	-	- 1,1
41 Berlin	17,0	57,3	61,6	- 0,63	0,5
43 Tel Aviv	-	- 48,7	0,9	-	1,0
44 Svanvik	46,0	53,7	52,0	- 0,20	0,2
45 Chaumont	35,0	30,3	28,6	- 1,27	- 1,5
46 London	-	10,0	4,2	-	- 1,3
47 Los Angeles	-	58,7	74,2	-	- 0,1
49 Antverps	-	51,7	36,4	-	- 3,0
50 Katowice	-	-	35,0	-	- 3,1

* the results of 1 year exposure from long-term exposure programme
marked values are not reliable due to not specified effects

Figure 2 - The weight changes of carbon steel samples after exposure in open atmosphere during periods 1996/97, 1997/98 and 2000/01

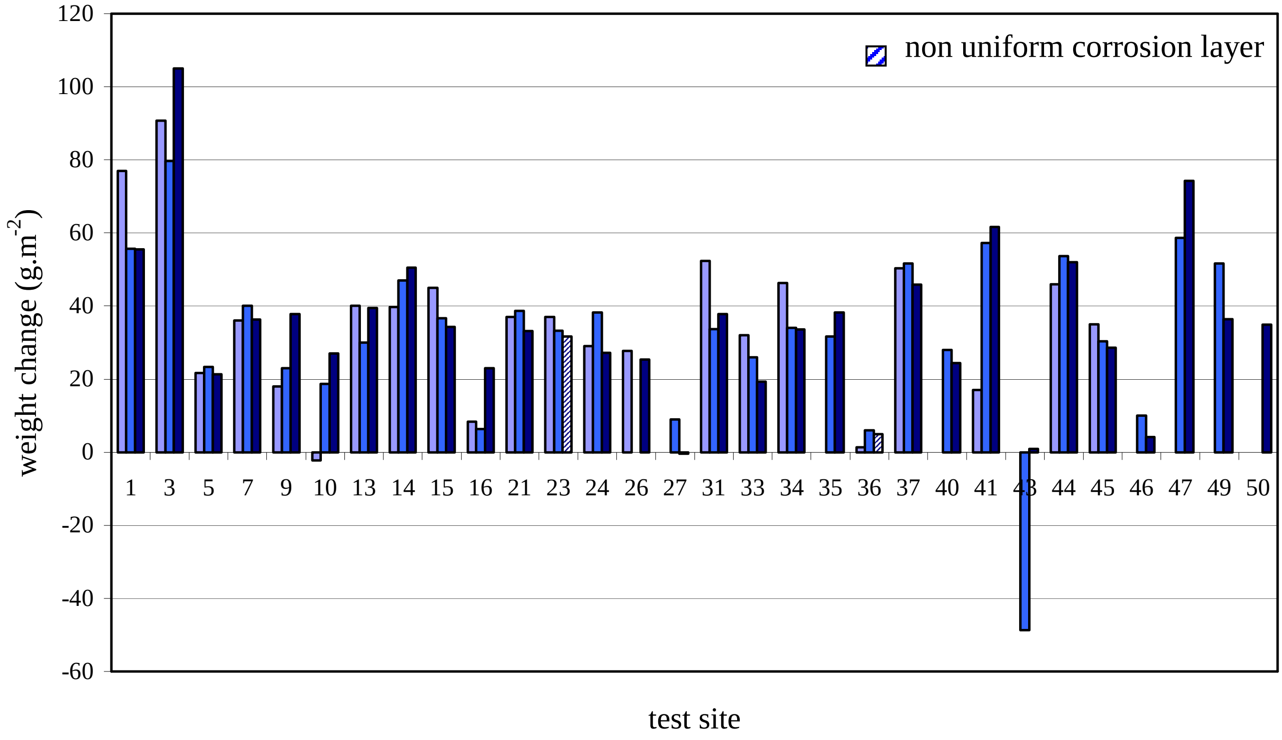
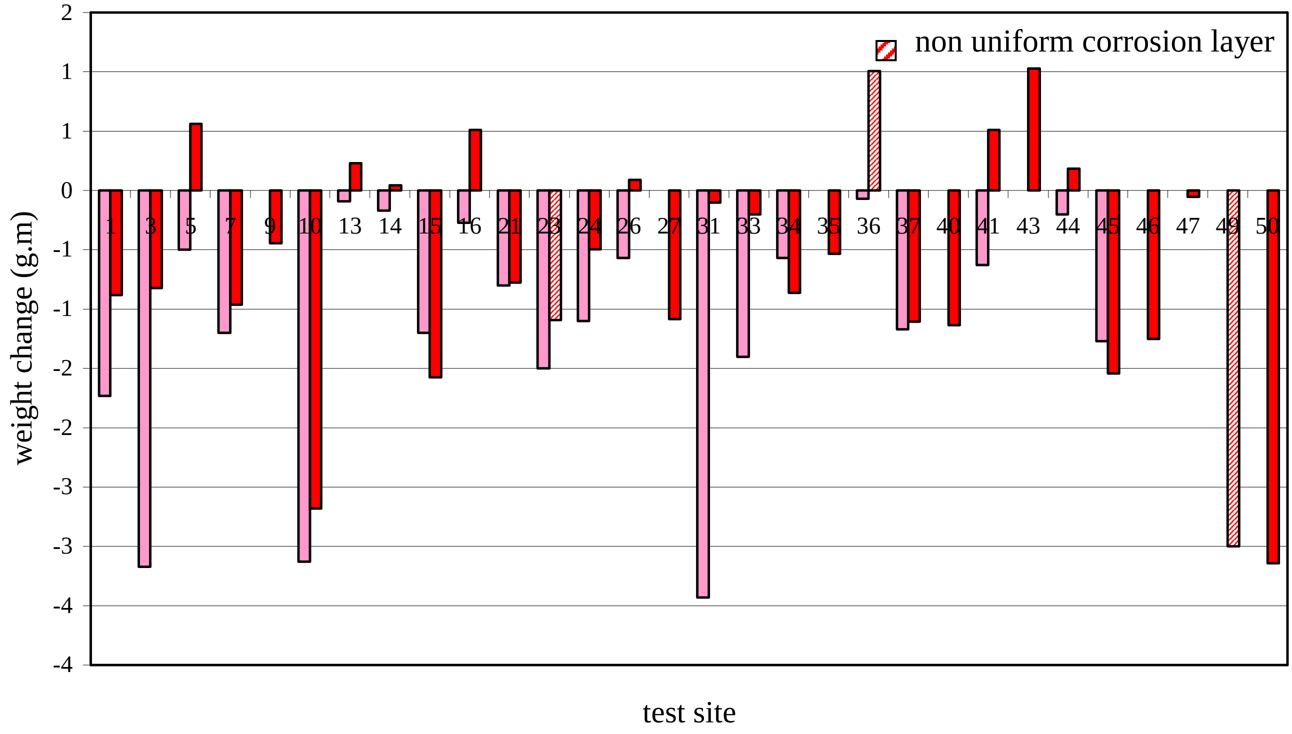


Figure 3 - The weight changes of zinc samples after exposure in open atmosphere during periods 1996/97 and 2000/01



2.3. Corrosion losses of carbon steel and zinc

Corrosion losses for individual samples of unalloyed steel and zinc exposed in open atmosphere evaluated for 1-year exposure (1996/97) are summarised in Tables 3 and 5 and presented in Figure 4. After 1 year of exposure in this period test sites with the greatest corrosion losses of samples were No. 3 (Kopisty) and No. 10 (Bottrop) for carbon steel and zinc and No. 36 (Lisbon) for carbon steel only.

Corrosion losses for individual samples of unalloyed steel and zinc exposed in open atmosphere evaluated for 1-year exposure in period 2000/01 are summarised in Tables 4 and 6 and presented in Figure 4. After 1 year of exposure in this period test sites with the greatest corrosion losses of samples were No. 50 (Katowice), No. 43 Tel Aviv, No. 10 (Bottrop) and No. 3 (Kopisty) for carbon steel and zinc, too.

In comparison period 1996/97 to period 2000/01 the significant decreasing of corrosion loss of carbon steel and zinc was found for 6 test sites: No. 1, 3, 9, 13, 14 and 31. The corrosion loss remained on the same level for 3 test sites: No. 10, 33 and 41. The increasing of corrosion loss was evaluated for 3 test sites: No. 16, 21 and 37. The corrosion loss for other test site did not show any significant and characteristics changes.

Table 3 - Corrosion losses (g/m^2) of unalloyed steel after 1 year´s exposure in open atmosphere (period 1996/97)

Test site	41	42	43	average
original test sites				
1 Prague	230,30	230,97	234,73	231,99
3 Kopisty	298,93	280,26	298,87	292,69
5 Ahtari	54,20	54,27	53,56	54,01
7 Waldhof-Langenbrugge	157,53	151,77	157,23	155,51
9 Langenfeld	201,69	197,59	217,57	205,61
10 Bottrop	296,88	290,14	300,01	295,68
13 Rome	109,54	109,25	107,14	108,64
14 Casaccia	133,10	136,08	135,69	134,62
15 Milan	151,18	148,94	146,69	148,93
16 Venice	187,45	184,53	191,78	187,92
21 Oslo	99,53	98,14	99,53	99,07
23 Birkenes	116,70	116,01	107,67	113,46
24 Stockholm South	110,04	105,20	104,68	104,12
26 Aspvreten	73,93	71,00	62,50	69,14
27 Lincoln Cathedral	-	-	-	-
31 Madrid	155,76	160,54	161,62	159,31
33 Toledo	36,10	36,16	36,26	36,17
34 Moskow	123,42	123,14	122,65	123,07
36 Lisbon	306,78	307,90	309,51	308,06
37 Dorset	129,80	130,43	128,49	129,54
new test sites				
40 Paris	-	-	-	-
41 Berlin	171,49	177,83	173,29	174,20
42 Athens	-	-	-	-
43 Tel Aviv	-	-	-	-
44 Svanvik	160,23	160,00	160,86	160,37
45 Chaumont	93,54	93,60	93,05	94,40
46 London	-	-	-	-
47 Los Angeles	-	-	-	-
49 Antverps	-	-	-	-

marked value is not included in calculated average

Table 4 - Corrosion losses (g/m^2) of unalloyed steel after 1 year´s exposure in open atmosphere (period 2000/01)

Test site	71	72	73	average
original test sites				
1 Prague	146,05	134,20	133,27	137,84
3 Kopisty	237,18	218,35	215,33	223,62
5 Ahtari	49,01	50,13	54,01	51,05
7 Waldhof-Langenbrugge	149,17	149,17	145,86	148,07
9 Langenfeld	105,34	98,84	98,00	100,73
10 Bottrop	295,60	294,03	289,41	293,01
13 Rome	87,27	76,15	76,34	79,92
14 Casaccia	87,05	-	109,86	98,46
15 Milan	183,15	184,96	183,69	183,94
16 Venice	151,28	147,33	147,61	148,74
21 Oslo	101,20	96,34	94,65	97,40
23 Birkenes	105,61	118,45	118,98	114,35
24 Stockholm South	118,96	114,26	116,12	116,45
26 Aspvreten	72,58	68,78	66,63	69,33
27 Lincoln Cathedral	185,30	199,25	201,63	195,40
31 Madrid	76,40	77,58	77,56	77,18
33 Toledo	47,29	45,93	47,66	46,96
34 Moskow	131,38	140,59	134,86	135,61
35 Lahemaa	92,08	91,03	101,34	94,82
36 Lisbon	229,47	224,23	223,11	225,60
37 Dorset	95,43	98,26	103,66	99,11
new test sites				
40 Paris	133,47	165,35	131,35	143,39
41 Berlin	168,78	168,65	184,56	174,00
43 Tel Aviv	261,72	239,76	267,84	256,44
44 Svanvik	153,12	145,78	144,97	147,96
45 Chaumont	63,72	54,45	57,23	58,47
46 London	164,36	159,47	185,46	169,76
47 Los Angeles	162,56	155,50	157,49	158,52
49 Antverps	193,60	179,57	182,46	185,21
50 Katowice	280,19	261,74	269,89	270,61

marked values are not reliable due to not specified effects

Table 5 - Corrosion losses (g/m^2) of zinc after 1 year´s exposure in open atmosphere (period 1996-1997)

Test site	41	42	43	average
1 Prague	5,32	5,58	5,99	5,63
3 Kopisty	8,74	8,78	8,72	8,75
5 Ahtari	3,08	3,05	3,06	3,06
7 Waldhof-Langenbrugge	3,08	3,56	3,78	3,47
9 Langenfeld	5,60	6,00	5,43	5,68
10 Bottrop	8,33	9,11	8,32	8,59
13 Rome	3,58	3,56	4,14	3,76
14 Casaccia	3,08	3,10	3,09	3,09
15 Milan	4,58	4,35	4,19	4,38
16 Venice	4,18	3,97	4,18	4,11
21 Oslo	2,12	2,39	2,46	2,32
23 Birkenes	3,63	3,52	3,35	3,50
24 Stockholm South	3,26	3,17	3,02	3,15
26 Aspvreten	2,51	2,41	2,95	2,62
27 Lincoln Cathedral	-	-	-	-
31 Madrid	2,37	2,39	-	2,38
33 Toledo	1,98	2,30	2,41	2,23
34 Moskow	4,24	4,16	3,98	4,13
36 Lisbon	4,26	3,87	-	4,07
37 Dorset	2,37	2,68	2,66	2,57
new test sites				
40 Paris	-	-	-	-
41 Berlin	6,44	-	6,22	6,33
42 Athens	-	-	-	-
43 Tel Aviv	-	-	-	-
44 Svanvik	2,95	2,94	2,63	2,84
45 Chaumont	3,16	2,62	3,24	3,01
46 London	-	-	-	-
47 Los Angeles	-	-	-	-
49 Antverps	-	-	-	-

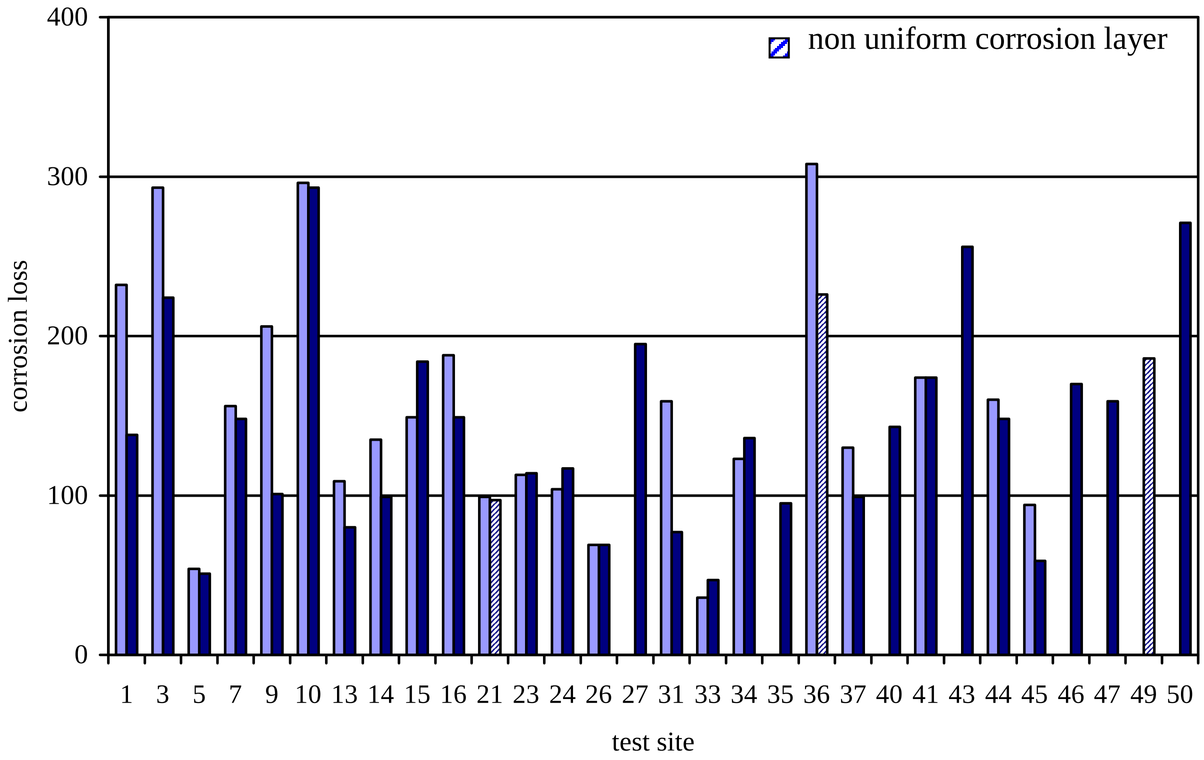
Table 6 - Corrosion losses (g/m^2) of zinc after 1 year´s exposure in open atmosphere (period 2000-2001)

Test site	71	72	73	average
1 Prague	3,98	4,00	3,59	3,86
3 Kopisty	4,48	4,72	4,72	4,64
5 Ahtari	6,17	5,71	5,42	5,77
7 Waldhof-Langenbrugge	4,52	4,84	4,32	4,56
9 Langenfeld	6,15	5,43	5,67	5,75
10 Bottrop	8,91	8,94	9,62	9,16
13 Rome	3,01	3,05	3,23	3,10
14 Casaccia	2,52	2,54	2,36	2,47
15 Milan	-	3,79	4,60	4,19
16 Venice	4,44	4,48	5,00	4,64
21 Oslo	3,41	3,45	3,48	3,45
23 Birkenes	8,65	8,52	8,26	8,48
24 Stockholm South	4,70	4,13	4,40	4,41
26 Aspvreten	3,57	3,44	3,48	3,50
27 Lincoln Cathedral	5,02	5,05	5,59	5,22
31 Madrid	2,27	2,05	1,92	2,08
33 Toledo	2,09	2,40	2,37	2,28
34 Moskow	6,87	6,85	6,53	6,75
35 Lahemaa	5,21	4,91	5,17	5,09
36 Lisbon	8,09	8,60	7,39	8,03
37 Dorset	3,65	3,82	3,72	3,73
new test sites				
40 Paris	4,93	5,00	4,77	4,90
41 Berlin	6,08	5,55	5,75	5,80
43 Tel Aviv	-	6,61	8,59	7,60
44 Svanvik	3,71	3,98	4,12	3,94
45 Chaumont	5,01	4,77	5,10	4,96
46 London	6,19	6,08	-	6,14
47 Los Angeles	3,95	5,99	4,98	4,97
49 Antverps	8,14	8,10	8,20	8,15
50 Katowice	9,70	10,03	9,89	9,88

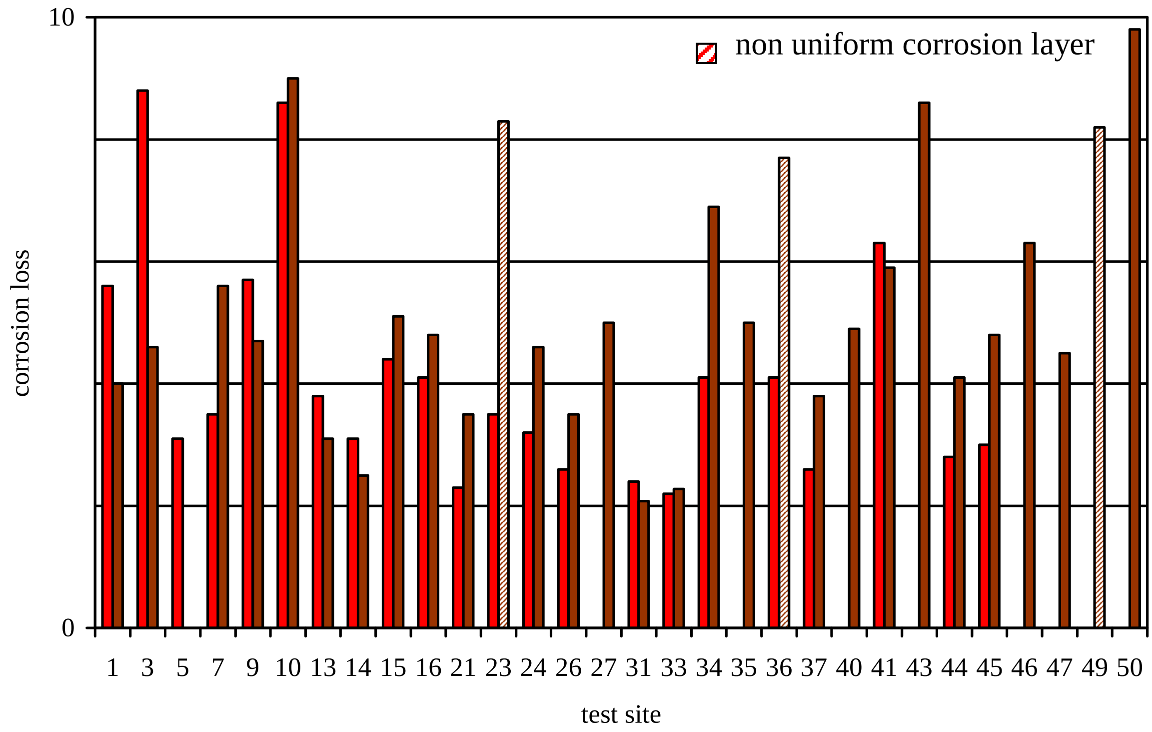
marked values are not reliable due to not specified effects

Figure 4 – Corrosion loss (g.m^{-2}) after 1 year of exposure in periods 1996/97 and 2000/01

carbon steel



zinc



2.4. Analysis of corrosion products

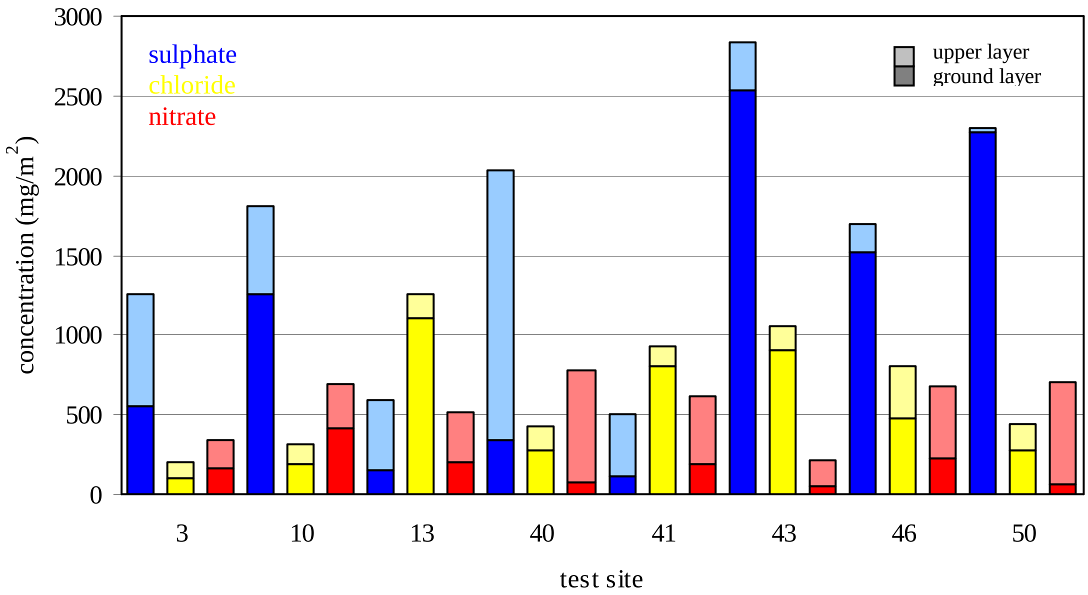
The concentrations of soluble ions (sulphates, chlorides and nitrates) were evaluated in water extract of corrosion products of selected samples of carbon steel and zinc exposed in period 2000/01 (test sites with relatively highest pollution of environment only). The results are in Figure 5.

The corrosion products from carbon steel had been got by mechanical removal of rust layer from a part of sample (about ¼ of area of sample) which had been extracted by demineralized water for 24 hours. Than the part of sample with adhesive corrosion layer had been immersed in deionized water for 24 hours. The two analysis had been done. The corrosion products from zinc had been get by immersion a part of sample (about ¼ of area of sample) in deionized water for 24 hours. The concentration of soluble ions in extracts was determinated by spectrophotometric method.

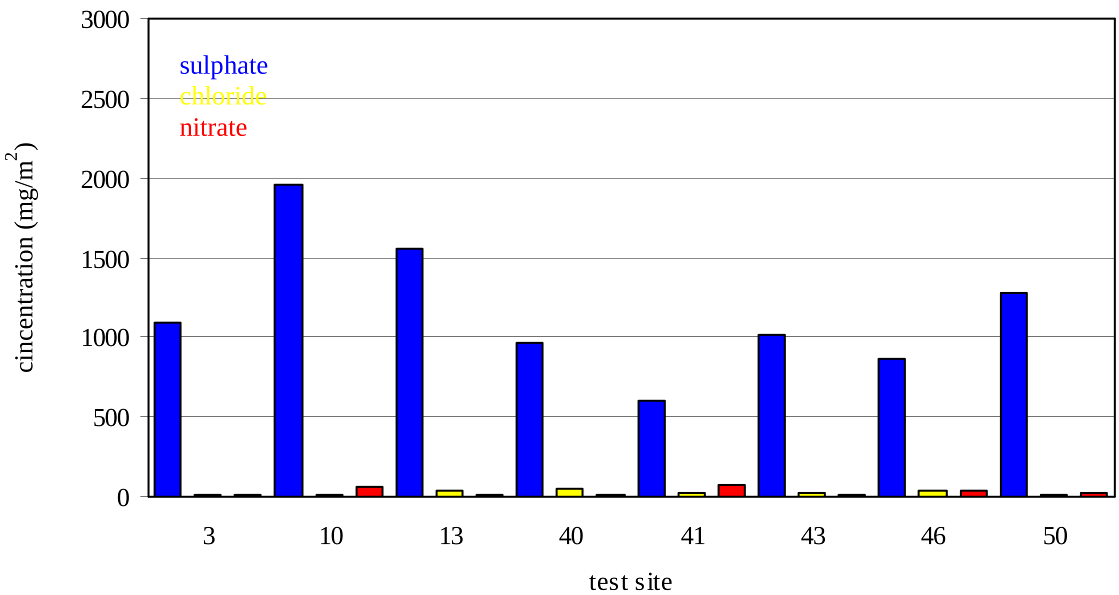
The concentrations of soluble ions are much higher in corrosion products of carbon steel than zinc (the mechanism of corrosion of metals is different and the thickness of layer of corrosion products is higher). Sulphates had been found as the dominant compound in corrosion products of each samples. The concentrations of other ions are significant in case of corrosion product of carbon steel only. The obtained results of composition of corrosion products of carbon steel and zinc very good correspond with dominant environmental pollution on test site. Significant concentration of chlorides had been found in corrosion products from test sites: No. 13 (Rome), No. 41 (Berlin), No. 43 (Tel Aviv) and No. 46 (London). Significant concentration of nitrates had been found in corrosion products from test sites: No. 10 (Bottrop), No. 41 (Berlin), No. 46 (London) and No. 50 (Katowice). Comparison of results obtained with the pollution characteristics of atmosphere will represent the next step of evaluation in future.

Figure 5 - Concentration of soluble ions in layers of corrosion products (period 2000/01)

carbon steel



zinc



3. Data treatment and evaluation of results

3.1.Environmental characteristics

The climatic parameters did not change on any test site significantly between these two yearly periods. There are very small differences typical for these parameters during long-term study. There is only one exception – test site No. 33 (Toledo), where yearly average temperature and relative humidity is different (14,0^{0}C resp. 12,2^{0}C and 59% resp. 71%). There is a difference between these two exposure periods in amount of precipitation for test site No. 43 Tel Aviv – 485 mm in 1997/98, resp. 254 mm in 2000/01.

During the exposure programme (19987-2001) the gaseous pollution at atmospheres in Europe dramatically changed. SO_2 concentration decreased so significantly that the dominant pollution there are NO_x, mainly in urban atmospheres – Figures 6 and 7. From Figure 7 it is obvious that the only test site with dominate influence of SO_2 is No 50 Katowice. In other test sites the concentration of NO_x is higher than SO_2.

There are the basic measured environmental parameter for the last two exposure periods in Tables 7 and 8. The comparison of yearly SO_2 concentration is in Table 9.

On many test sites the pollution situation also did not change with exceptions:

- significant decreasing of SO_2 on test sites No. 1, 10, 21, 31, 43 and 49
- significant decreasing of NO_x on test sites No. 15, 21, 31, 33 and 43

Test site with the greatest changes in air pollution between mentioned two yearly periods is No. 43 Tel Aviv – see Tables 7 and 8.

The test sites with high effect of NO_x belong No. 15 (although concentration decreased slowly) and Nos. 40, 41, 46 and 49. In period 2000/01 on test sites No. 40 and 49 NO_x concentration decreased too. There was found significant difference between yearly NO_x concentration at test site No 40 for the last two exposure periods – 70 $\mu g.m^{-3}$, resp. 46 $\mu g.m^{-3}$.

In frame of multi-pollutant programme the ozone concentration was included among the mandatory measured pollution parameters. The average yearly concentration had been relatively high – from 20 $\mu g.m^{-3}$ to 90 $\mu g.m^{-3}$. Test site with the lowest values was No. 36 Lisbon. Test sites with the highest values were No. 33 and 45.

Table 7 - Average yearly environmental parameters for exposure period 1997/98

Test site	CLIMATE			GASES			PRECIPITATION					
	Temp	Rh	Sun	SO_2	NO_2	O_3	mm	pH	SO_4^{2-}	NO_3^-	Cl^-	Cond
	^0C	%	MJ/m^2	$\mu g/m^3$	$\mu g/m^3$	$\mu g/m^3$			mg/l	mg/l	mg/l	µS/cm
1 Prague	9,9	76	2971	15,3	23,7	48	522	5,56	11,34	2,19	1,95	27,6
3 Kopisty	9,9	76	3921	18,3	32,6	55	420	4,62	34,03	2,77	1,73	44,6
5 Athari	3,5	80	2889	0,9	3,0	60	742	4,74	0,25	0,21	0,15	10,7
7 Waldhof-Langenbrugge	9,5	83	3104	2,1	8,7	48	786	5,04	0,64	0,57	0,73	17,6
9 Langenfeld	10,9	80	2494	8,3	33,5	33	930	4,92	0,89	0,57	1,55	
10 Bottrop	11,5	81	2531	24,6	38,2	30	1044	4,84	0,99	0,46	1,20	22,1
13 Rome	19,4	65	4360	3,7	37,8	33	1125					
14 Casaccia	14,5	74	5178	5,2	21,0	30						
15 Milan	14,5	69	4940	15,4	83,9	38	1077					
16 Venice	13,5	83	4999	7,4			742	6.10				
21 Oslo	6,6	79	2521	4,1	27,5	36	523	5,20	0,85	0,55	0,87	20,7
23 Birkenes	6,2	79	2626	0,2	1,1	55	1744	4,50	0,61	0,47	1,50	25,5
24 Stockholm South	6,7	76	3048	2,6	20,3	44	463	4,63	0,54	0,38	0,48	17,1
26 Aspvreten	5,9	86	3301	0,6	2,9	51	479	4,59	0,41	0,37	0,57	17,3
27 Lincoln Cathedral	10,2	81	3224	8.4	19.1	51	708	4,61	1,64	0,56	3,56	
31 Madrid	12,9	61	5722	11,8	22,1	56	765	6,05	0,77	0,37	1,03	16,0
33 Toledo	14,0	59	5905	1,5	11,3	89	872	5,80	0,56	0,26	0,92	10,7
34 Moscow	6,5	74		31.5	28,0	42		6,68	1,42		1,57	45,8
35 Lahemaa	5,4	82	3238	0.5	0,7	55	859	5,16	1,64	0,24	0,78	19,4
36 Lisbon	17,9	63		17,7	42,0	12	252	5,98	13,30	5,02	13,30	75,3
37 Dorset	7,4	75	4435	2,4	9,7	62	788	4,31	0,76	0,52	0,11	24,2
40 Paris	13,4	67	4250	14,2	70,0	31	572	5,71	1,81	0,72	2,47	43,7
41 Berlin	10,4	77	3113	10,9	37,7	22	486		6,98	4,34	2,09	
43 Tel Aviv	24,6	83		35,0	38,3	40	485	5.64	0.47	0.44	3.53	
44 Svanvik	0,2	80	1967	7,5	0,9	54	344	4,77	0,57	0,12	1,87	20,7
45 Chaumont	6,9	77	4388	1,3	7,7	86	1053	4,99	0,27	0,21	0,18	9,4
46 London	12,2	70	3228	6.3	45.3	36	706	5,65	0,54	0,26	4,07	
47 Los Angeles	17,4	61		0,6	21,7	48		5.77				
49 Antverps	11,4	76	3027	22,8	52,8	28		5.07	1.46	0.47	3.86	35.6

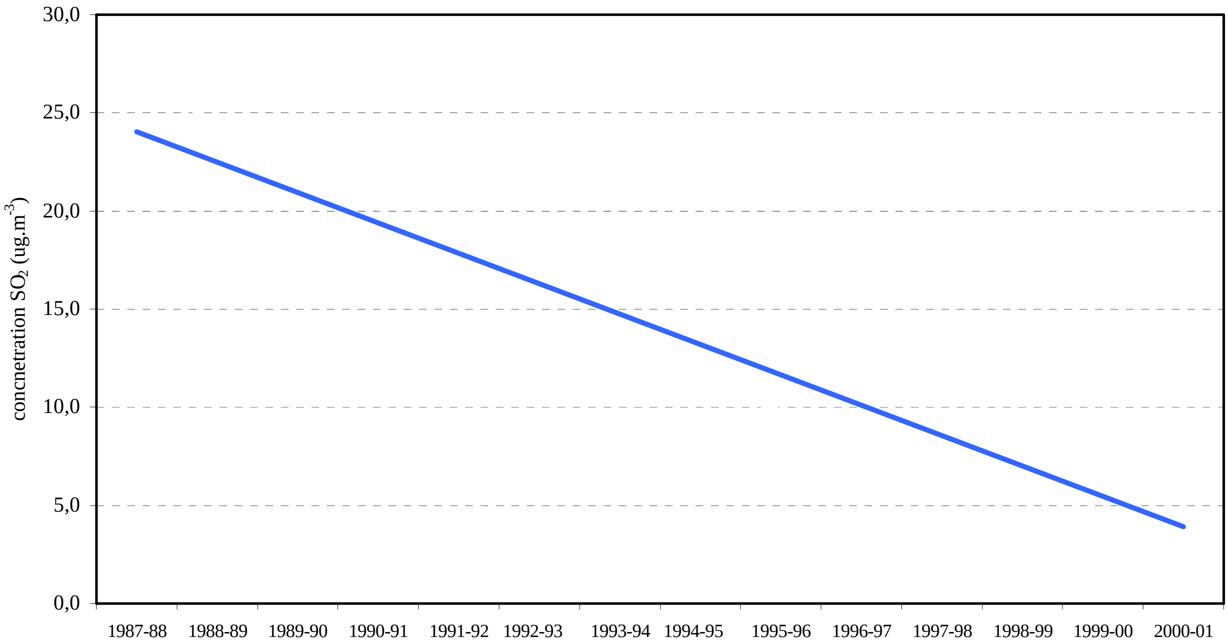
Table 8 - Average yearly environmental parameters for exposure period 2000/01

Test site	CLIMATE			GASES			PRECIPITATION					
	Temp ^0C	Rh %	Sun MJ/m^2	SO_2 $\mu g/m^3$	NO_2 $\mu g/m^3$	O_3 $\mu g/m^3$	mm	pH	SO_4^{2-} mg/l	NO_3^- mg/l	Cl^- mg/l	Cond $\mu S/cm$
1 Prague	9,5	79	2992	8,8	21,7	41	601	6,59	5,79	2,45	1,99	40,9
3 Kopisty	9,2	80	3983	17,9	24,8	40	510	4,41	9,87	4,55	1,70	47,6
5 Athari	4,8	82	3072	0,8	3,1	57	845	4,78	0,26	0,20	0,13	10,0
7 Waldhof-Langenbrugge	9,4	81	3474	2,3	8,4	51	620	4,85	0,45	0,50	0,64	17,2
9 Langenfeld	11,6	79	2895	5,0	30,5	36	997	4,89	1,06	0,50	0,77	21,4
10 Bottrop	11,7	81	2907	17,9	32,6	33	791	4,80	2,09	0,62	1,54	31,2
13 Rome	17,9	66			24,1	35						
14 Casaccia	13,4	64		6,4								
15 Milan	15,9	71		12,9	72,8	40						
16 Venice	14,9	83		7,8								
21 Oslo	7,2	75	2617	3,1	20,8	36	1050	4,72	0,42	0,41	0,73	16,6
23 Birkenes	6,6	83	2706	0,3	1,8	53	2333	4,59	0,47	0,44	1,84	23,2
24 Stockholm South	8,1	81	3044	1,9	17,5	47	635	4,75	0,46	0,34	0,40	16,7
26 Aspvreten	7,2	86	3379	0,6	2,7	57	772	4,39	0,58	0,48	0,69	23,8
27 Lincoln Cathedral	9,7	81	3900	7,5	22,6	49	831					
31 Madrid	15,0	62	4605	1,2	11,0	54	560	6,33	0,59	0,32	1,18	13,3
33 Toledo	12,2	71	5919	1,2	3,1	92	739	6,44	0,69	0,31	1,62	16,1
34 Moscow	7,4	69					812					
35 Lahemaa	6,9	81	3446	1,3	2,2	60	668	4,69	0,39	0,18	0,41	12,0
36 Lisbon	13,8	78										
37 Dorset	6,5	78			7,8	50		4,09				
40 Paris	12,7	74	3857	10,1	45,6	35	731	5,17	1,19	1,14	1,82	35,7
41 Berlin	11,1	82	3514	9,8	44,2	29			2,84	2,93	0,72	
43 Tel Aviv	22,0	70		6,6	17,8	24	254					
44 Svanvik	1,0	80		5,2	1,6	49	361	4,89	0,75	0,15	1,22	22,4
45 Chaumont	7,2	80	4128	1,0	6,9	82	1281	5,13	0,23	0,20	0,13	7,4
46 London	12,1	69	2364	5,8	44.0	35	907					
47 Los Angeles	16,4	68	6336									
49 Antverps	11,7	75	3510	13,5	47,2	30	993	4,75	0,97	0.41	2.37	25,9
50 Katowice	9,4	81	1582	34,4	22,7	56	870	4,41	3,93	1,97	1,35	33,9

Table 9 – Average yearly concentration of SO_2

Test site	1988	1989	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2001
1 Prague	77,5	74,2	58,1	61,4	43,7	41,2	40,2	32,1	31,5	23,6	15,3	13,1	12,6	8,8
3 Kopisty	83,3	94,6	78,4	75,9	57,8	49,0	49,5	49,2	41,1	30,6	18,3	17,8	16,0	17,9
5 Ahtari	6,3	5,3	1,8	1,8	0,8	0,9	1,3	0,8	1,0	0,6	0,9	0,9	0,5	0,8
7 Waldhof-Langenbrugge	13,7	11,4	11,0	12,9	7,3	8,2	7,8	3,9	5,9	2,9	2,1	1,5	0,9	2,3
9 Langenfeld	24,5	25,7	20,3	23,8	19,9	16,3	13,6	11,1	12,8	10,5	8,3	6,5	5,3	5,0
10 Bottrop	50,6	48,6	48,5	53,0	53,5	41,6	35,5	30,2	33,8	29,4	24,6	20,4	18,3	17,9
13 Rome	29,4	44,9	38,5	24,4	2,4	6,8	14,4	5,8			3,7			
14 Casaccia	8,3	8,3	7,4	6,4	4,7	7,5	4,7	5,2			5,2	3,0	3,4	6,4
15 Milan	72,2	82,7	65,4	50,3	58,5	39,4	32,4	22,1	12,2	14,6	15,3	20,1	15,1	12,9
16 Venice	21,1	25,7	20,2	16,4	18,6	11,0	7,1	6,3	3,0		7,4	5,5	6,4	7,8
21 Oslo	14,4	12,6	7,9	8,6	6,6	6,0	5,2	2,9			4,1	5,2	3,4	3,1
23 Birkenes	1,3	1,1	0,9	1,1	0,8	0,7	0,9	0,7	0,8	0,4	0,2	0,3	0,2	0,3
24 Stockholm South	16,6	12,6	8,4	6,3	5,7	5,7	5,4	4,2	4,6	3,3	2,6	3,2	2,2	1,9
26 Aspvreten	3,3	1,9	2,0	2,6	1,8	1,3	1,8	1,1	1,4	0,8	0,6	0,8	0,5	0,6
27 Lincoln Cathedral	17,7	19,6	15,5	20,2	20,4	21,2	7,8	8,4			8,4	9,8	8,0	7,5
31 Madrid	18,4	18,1	15,3	10,3	8,6	8,2	7,6	7,8	5,7	11,4	11,8	6,5	4,2	1,2
33 Toledo	3,3	8,6	13,5	6,0	4,6	1,7	3,5	4,2	3,9	1,1	1,5	2,3	1,9	1,2
34 Moskow	19,2	25,5	30,8	26,0	28,0	28,7	18,9	16,4			31,5	23,6		
35 Lahemaa	0,9	0,3	0,6	0,6	0,6	0,6	0,6	0,6	1,1	0,8	0,5	1,7	1,4	1,3
36 Lisbon	6,8	11,9	6,6	11,3	41,1	16,1	10,0	4,7	5,7		17,7	15,2	27,2	
37 Dorset	3,3	4,2	3,0	2,7	2,1	2,1	1,5	3,3	3,4	2,5	2,4	1,9		
40 Paris											14,2	11,1	11,1	10,1
41 Berlin										16,3	10,9	10,9	7,0	9,8
43 Tel Aviv											35,0	60,3	9,8	6,6
44 Svanvik									7,4		7,5	7,6	5,9	5,2
45 Chaumont										1,5	1,3	1,3	1,0	1,0
46 London										9,6	6,3	7,6	5,9	5,8
47 Los Angeles										1,5	0,6	0,4		
49 Antverps											22,8	16,5	16,8	13,5

Figure 6 – Trends in SO_2 concentration at 20 test sites in period 1987-2001



Detailed course of SO_2 concentration decreasing for test sites with the highest values (No. 1, 3, 10) and the lowest values (No. 5, 23, 35) in comparison to mean value

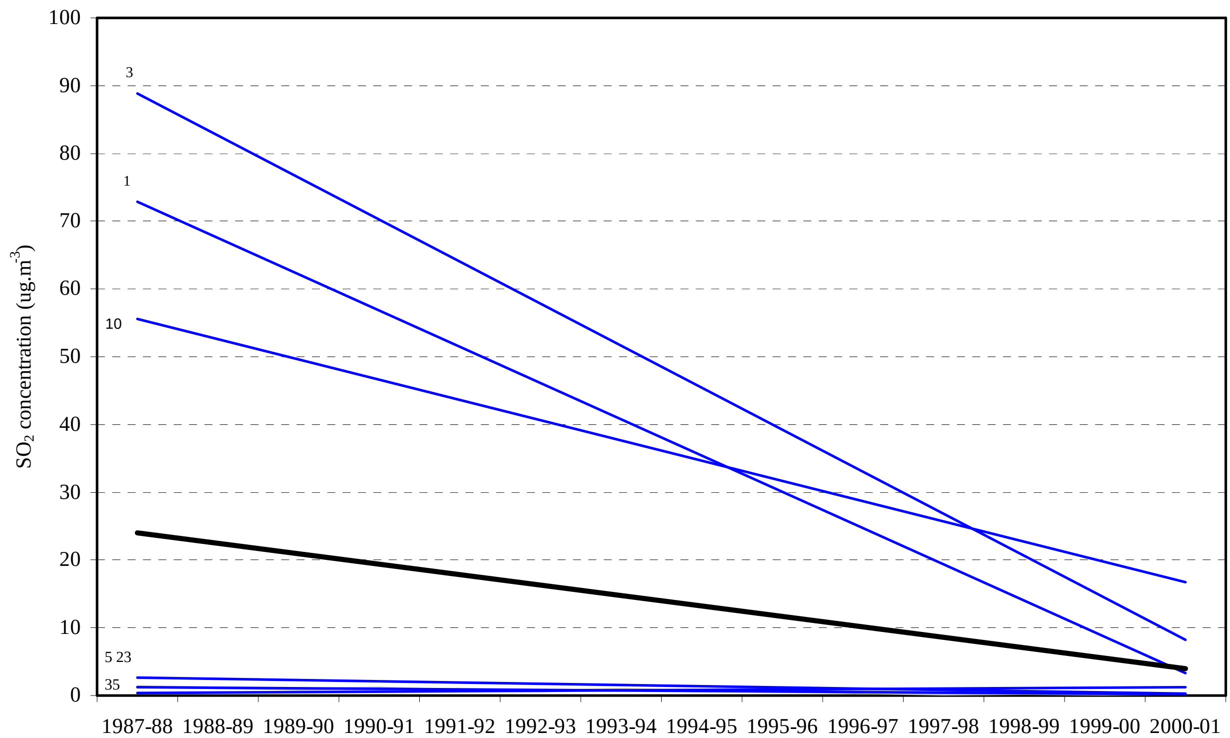
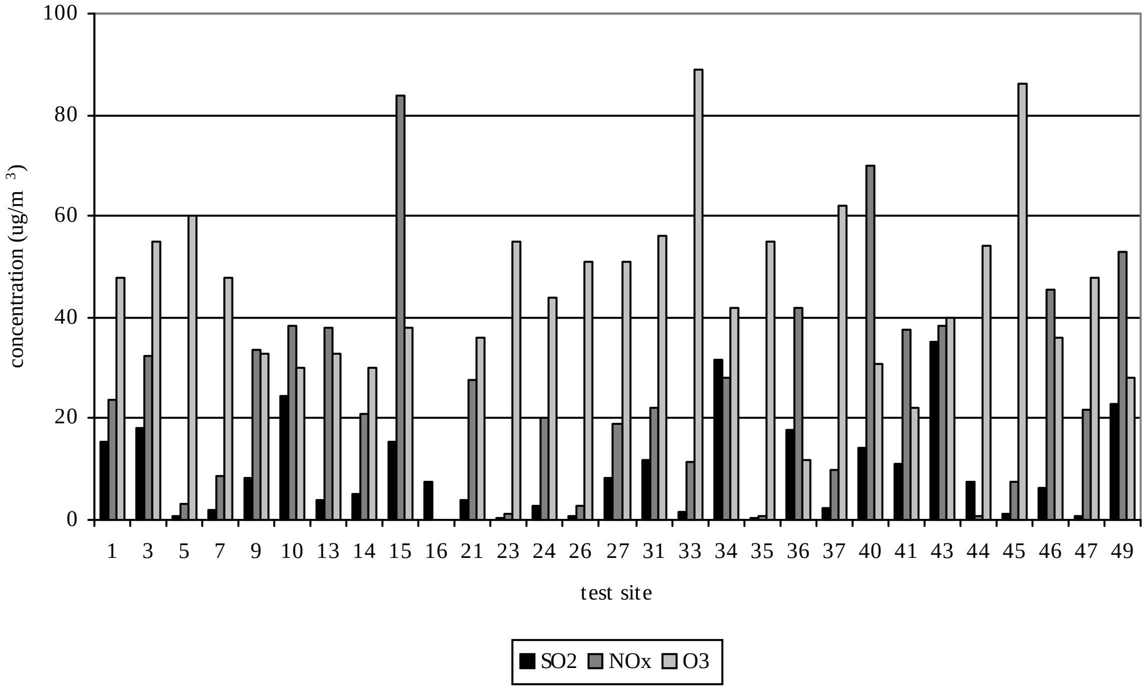
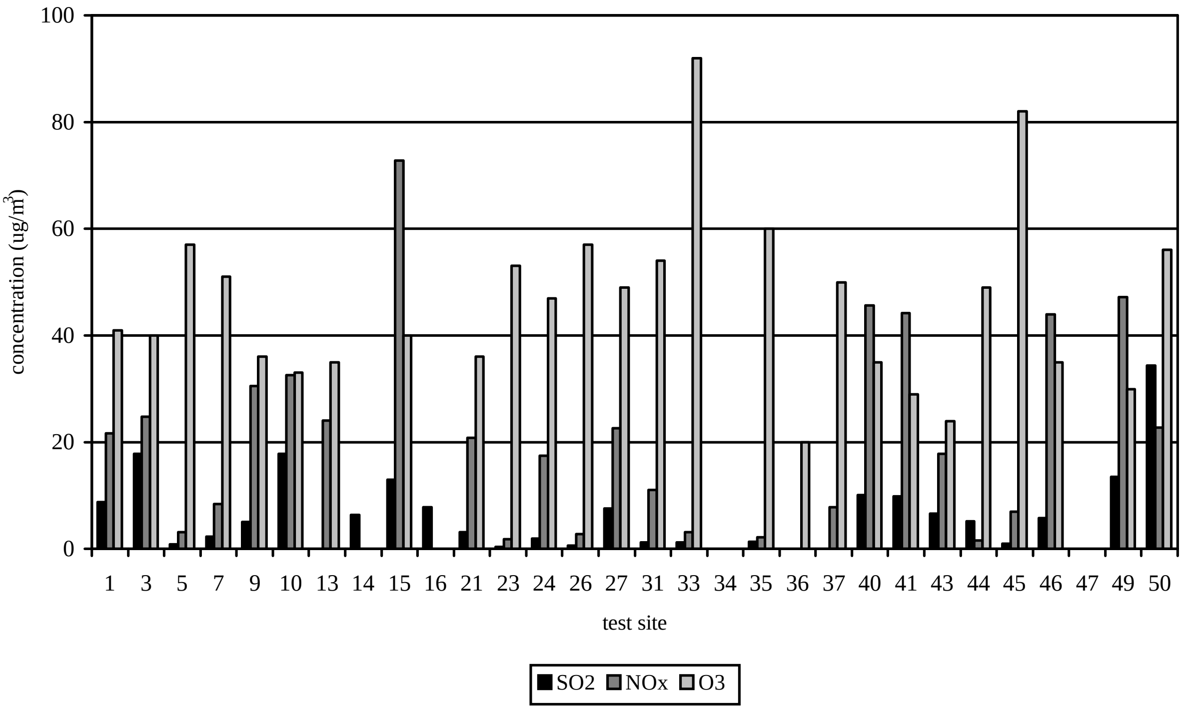


Figure 7 - Gaseous pollutants

Mean yearly average values in period 1997/99



Mean yearly average values in period 2000/01



3.2. Comparison of corrosivity of atmosphere in different periods

Samples of unalloyed carbon steel and zinc were exposed in the programme as standard specimens for determination of corrosivity of test sites (according to ISO 9226). Within the framework of the programme the exposures of these samples were repeated to validate decreasing trend of corrosivity due to decreasing of SO_2 pollution.

Comparisons of corrosion losses for different exposure periods are given in Tables 10 and 11 and Figures 8 and 9. There are not values for test sites No 27, 40, 42, 43, 46, 47 and 49 (samples were not exposed in this period).

Corrosion losses of both metals significantly decreased. The decreasing trend is more evident for carbon steel than for zinc. The corrosion of carbon steel is more affected by SO_2 pollution; corrosion of zinc is more affected by climatic conditions, especial by wetness of surface of samples.

The decreasing is more significant for test sites with high pollution of SO_2 in period 1987/88. Pollution situation on test site No 36 Lisbon is different; test site was moved on the other location of building; restoration works on building can increase local corrosivity - corrosion loss of carbon steel is on the similar level in all exposure periods.

Table 10 - Comparison of corrosion losses (g/m^2) of unalloyed steel after 1 year´s exposure in open atmosphere in different exposure periods

Test site	period 1987-88	period 1992-93	Period 1994-95	period 1996-97	period 1997-98	period 2000-01
1 Prague	438 C4	271 C3	241 C3	232 C3	182 C2	138 C2
3 Kopisty	557 C4	350 C3	352 C3	293 C3	239 C3	224 C3
5 Ahtari	132 C2	48 C2	59 C2	54 C2	53 C2	51 C2
7 Waldhof-Langenbrugge	264 C3	231 C3	166 C2	156 C2	144 C2	148 C2
9 Langenfeld	293 C3	231 C3	210 C3	206 C3	204 C3	101 C2
10 Bottrop	373 C3	347 C3	294 C3	296 C3	311 C3	293 C3
13 Rome	178 C2	-	124 C2	109 C2	134 C2	80 C2
14 Casaccia	235 C3	-	148 C2	135 C2	125 C2	98 C2
15 Milan	366 C3	-	198 C2	149 C2	173 C2	184 C2
16 Venice	245 C3	-	212 C3	188 C2	211 C3	149 C2
21 Oslo	229 C3	135 C2	101 C2	99 C2	93 C2	97 C2
23 Birkenes	194 C2	132 C2	109 C2	114 C2	101 C2	114 C2
24 Stockholm South	264 C3	120 C2	103 C2	104 C2	125 C2	117 C2
26 Aspvreten	147 C2	75 C2	81 C2	69 C2	62 C2	69 C2
27 Lincoln Cathedral	315 C3	308 C3	237 C3	-	270 C3	195 C2
31 Madrid	222 C3	162 C2	151 C2	159 C2	72 C2	77 C2
33 Toledo	45 C2	26 C2	36 C2	36 C2	54 C2	47 C2
34 Moskow	181 C2	141 C2	121 C2	123 C2	135 C2	136 C2
35 Lahemaa	185 C2	-	-	-	106 C2	95 C2
36 Lisbon	224 C3	308 C3	204 C3	289 C3	214 C3	226 C3
37 Dorset	149 C2	110 C2	104 C2	120 C2	116 C2	99 C2
40 Paris	-	-	-	-	137 C2	143 C2
41 Berlin	-	-	-	174 C2	179 C2	174 C2
43 Tel Aviv	-	-	-	-	324 C3	256 C3
44 Svanvik	-	-	-	160 C2	166 C2	148 C2
45 Chaumont	-	-	-	94 C2	67 C2	58 C2
46 London	-	-	-	-	177 C2	170 C2
47 Los Angeles	-	-	-	-	136 C2	159 C2
49 Antverps	-	-	-	-	171 C2	186 C2
50 Katowice	-	-	-	-	-	271 C3

C2, C3 , C4 ..... corrosivity categories according ISO 9223

Reliability of marked values will be checked in connection with the next trend exposure (2002-03)

Table 11 - Comparison of corrosion losses (g/m^2) of zinc after 1 year´s exposure in open atmosphere in different exposure periods

Test site	period 1989-90	period 1992-93	period 1994-95	period 1996-97	period 2000-01
1 Prague	7,0 C3	7,7 C3	5,6 C3	5,6 C3	3,9 C2
3 Kopisty	11,5 C3	11,6 C3	12,1 C3	8,8 C3	4,6 C2
5 Ahtari	7,6 C3	6,6 C3	4,6 C2	3,1 C2	5,8 C3
7 Waldhof-Langenbrugge	7,8 C3	9,0 C3	4,2 C2	3,5 C2	4,6 C2
9 Langenfeld	6,6 C3	9,0 C3	7,6 C3	5,7 C3	5,8 C3
10 Bottrop	10,6 C3	15,2 C3	7,8 C3	8,6 C3	9,2 C3
13 Rome	9,7 C3	-	3,4 C2	3,8 C2	3,1 C2
14 Casaccia	9,9 C3	-	3,1 C2	3,1 C2	2,5 C2
15 Milan	12,1 C3	-	5,5 C3	4,4 C2	5,2 C3
16 Venice	7,6 C3	-	6,1 C3	4,1 C2	4,6 C2
21 Oslo	5,6 C3	6,6 C3	3,5 C2	2,3 C2	3,5 C2
23 Birkenes	8,4 C3	10,5 C3	5,0 C3	3,5 C2	8,5 C3
24 Stockholm South	6,0 C3	4,5 C2	4,2 C2	3,2 C2	4,4 C2
26 Aspvreten	6,7 C3	4,8 C2	6,0 C3	2,6 C2	3,5 C2
27 Lincoln Cathedral	12,3 C3	10,6 C3	7,0 C3	-	5,2 C3
31 Madrid	4,8 C2	3,5 C2	2,3 C2	2,4 C2	2,1 C2
33 Toledo	3,9 C2	4,7 C2	1,7 C2	2,2 C2	2,3 C2
34 Moskow	8,6 C3	6,5 C3	4,6 C2	4,1 C2	6,8 C3
35 Lahemaa					5,1 C3
36 Lisbon	-	10,4 C3	5,6 C3	4,1 C2	8,0 C3
37 Dorset	6,2 C3	5,2 C3	6,1 C3	2,6 C2	3,1 C2
40 Paris	-	-	-	-	4,9 C2
41 Berlin	-	-	-	6,3 C3	5,8 C3
43 Tel Aviv	-	-	-	-	7,6 C3
44 Svanvik	-	-	-	2,8 C2	3,9 C2
45 Chaumont	-	-	-	3,0 C2	5,0 C2
46 London	-	-	-	-	6,1 C3
47 Los Angeles	-	-	-	-	5,0 C2
49 Antverps	-	-	-	-	8,2 C3
50 Katowice					9,9 C3

C2, C3 , C4 ..... corrosivity categories according ISO 9223

Reliability of marked values will be checked in connection with the next trend exposure (2002-03)

Figure 8 – Comparison of corrosion losses (g.m^{-2}) of unalloyed carbon steel in different exposure periods

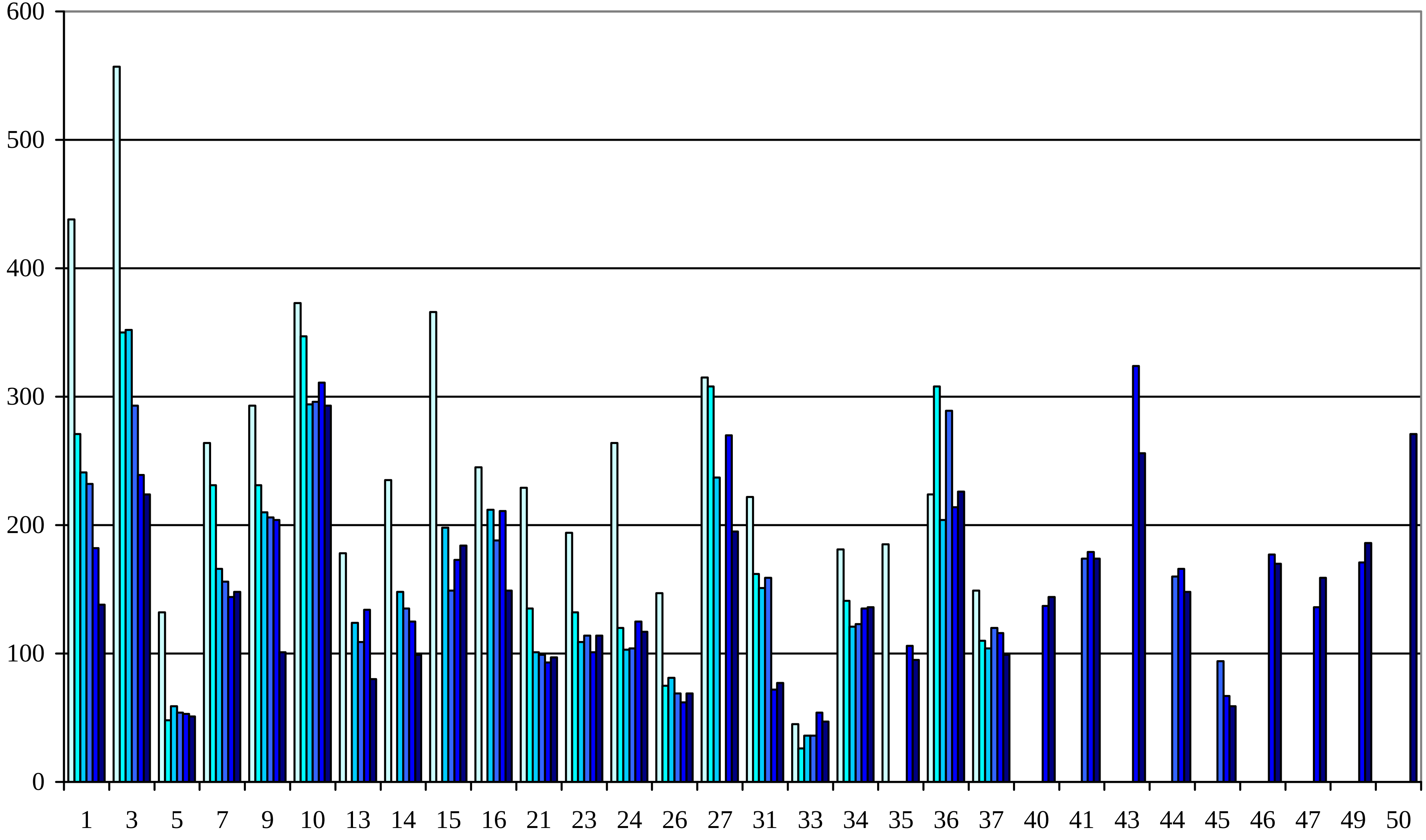
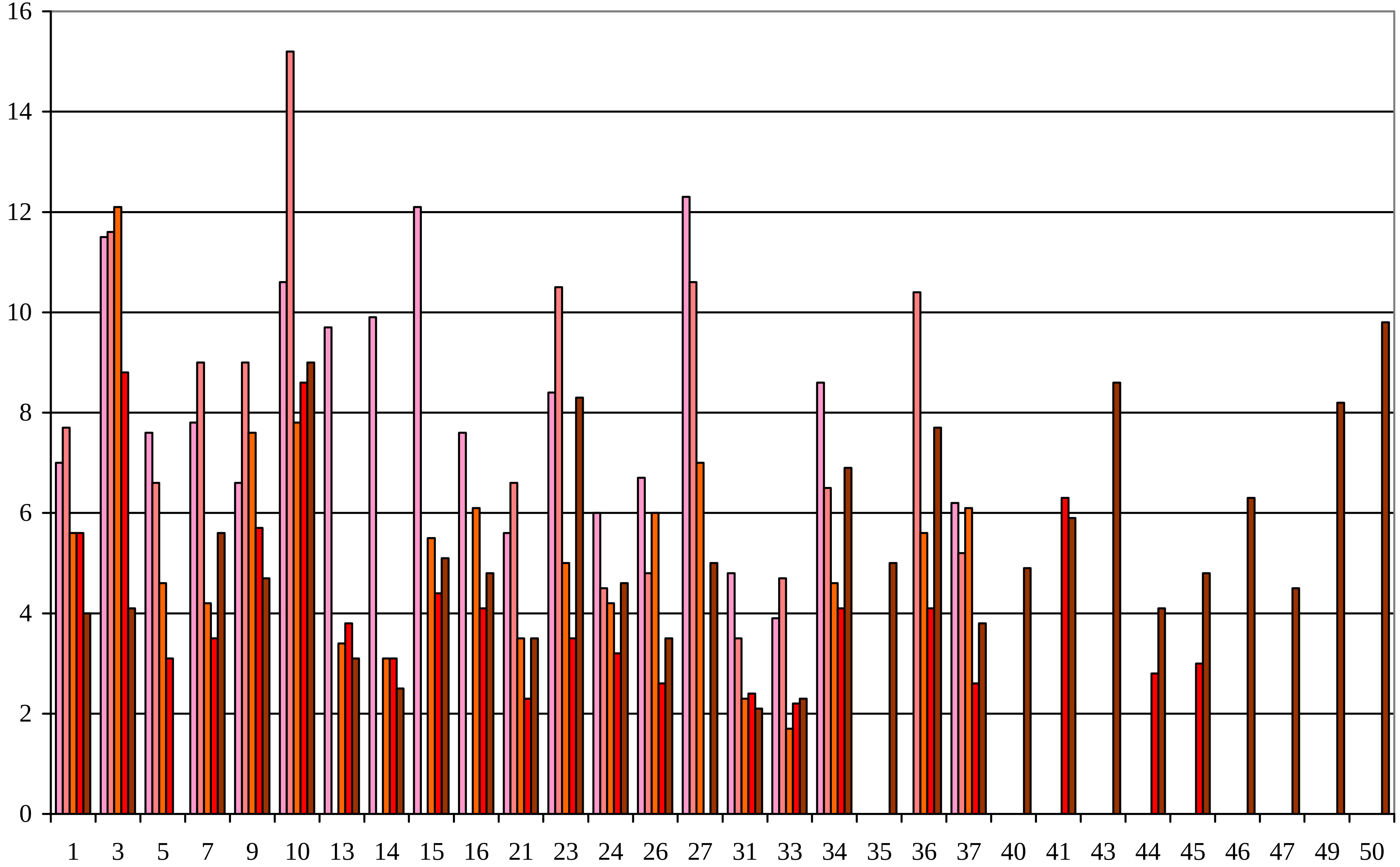


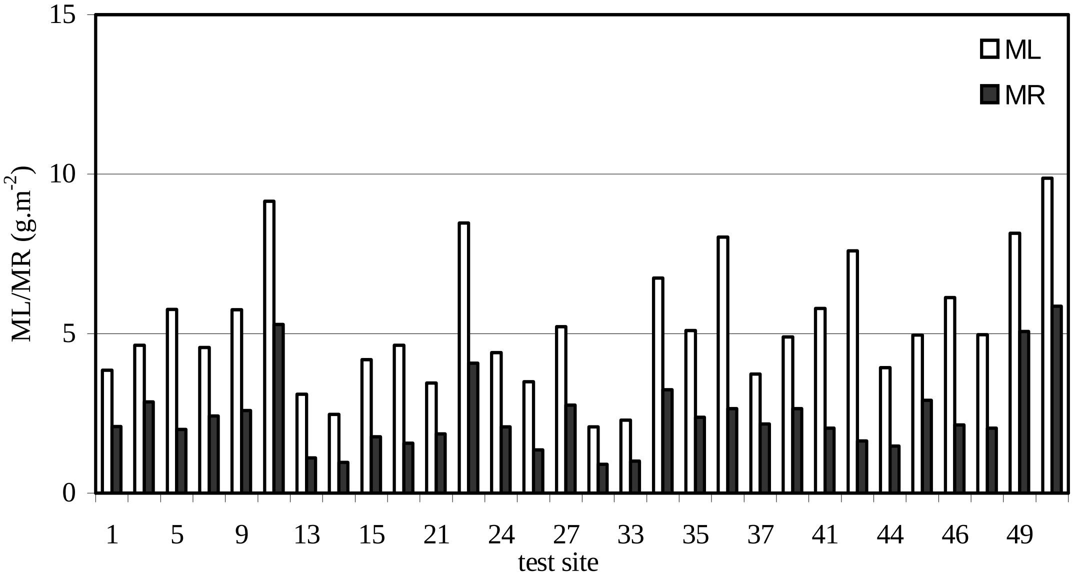
Figure 9 – Comparison of corrosion losses (g.m^{-2}) of zinc in different exposure periods



3.3. Runoff

After exposure of zinc trend samples in period 2000/01 the evaluation of runoff of zinc had been performed. The evaluation method - gravimetric method - had been elaborated in frame of this programme [1, 2]. The results are in Table 12 and Figure 10.

Figure 10 - Comparison of corrosion loss (ML) and runoff (MR) for period 2000/01



The obtained results for one-year exposure period good correspond with values presented by sub-centre for zinc - EMPA [2]. The average value of runoff of zinc had been found 2,4 $g.m^{-2}$ which represents about 46 % of yearly corrosion loss of zinc.

Table 12 - Corrosion loss (ML) and runoff (MR) of zinc in exposure period 2000/2001

Test site	ML_1	MR_1	
	(g/m^2)	(g/m^2)	(%)*
1 Prague	3,86	2,09	54,2
3 Kopisty	4,64	2,36	61,7
5 Ahtari	5,77	2,00	34,7
7 Waldhof-Langenbrugge	4,56	2,42	53,1
9 Langenfeld	5,75	2,59	45,0
10 Bottrop	9,16	5,30	57,9
13 Rome	3,10	1,11	35,8
14 Casaccia	2,47	0,97	39,2
15 Milan	4,19	1,76	42,0
16 Venice	4,64	1,57	33,9
21 Oslo	3,45	1,86	54,0
23 Birkenes	8,48	4,08	48,1
24 Stockholm South	4,41	2,08	47,2
26 Aspvreten	3,50	1,36	38,9
27 Lincoln Cathedral	5,22	2,76	52,9
31 Madrid	2,08	0,90	43,3
33 Toledo	2,28	1,01	44,2
34 Moskow	6,75	3,24	48,0
35 Lahemaa	5,09	2,38	46,7
36 Lisbon	8,03	2,65	33,0
37 Dorset	3,73	2,17	58,1
40 Paris	4,90	2,66	54,3
41 Berlin	5,80	2,04	35,2
43 Tel Aviv	7,60	1,64	21,6
44 Svanvik	3,94	1,48	37,6
45 Chaumont	4,96	2,92	58,9
46 London	6,14	2,15	35,0
47 Los Angeles	4,97	2,04	41,0
49 Antverps	8,15	5,07	62,2
50 Katowice	9,88	5,86	59,3

* expressed as a per cent of corrosion loss

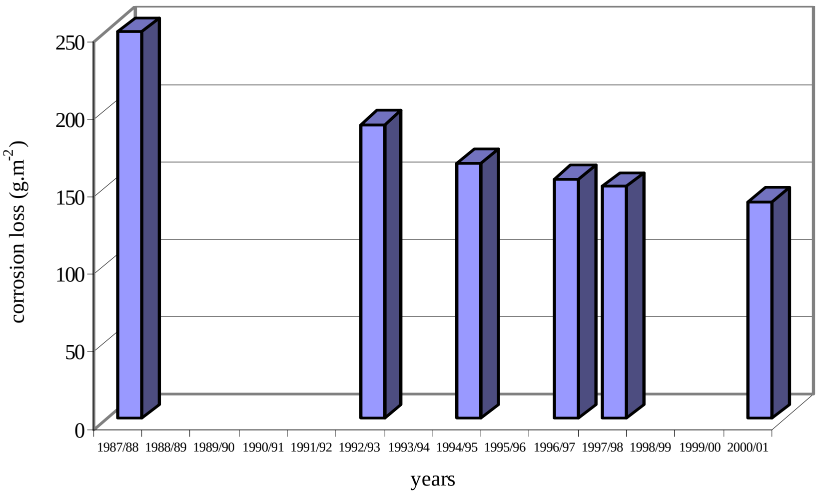
3.4. Analysis of results

The trend analysis has indicated that the pollution reduction has been smaller in the second phase of the programme. The comparison of gaseous pollution shows a dramatic reduction in the SO_2 concentration during period 1987-1998 – in average the reduction is close to 75%. There were found reduction about 40% for NO_x and no reduction for O_3. In the period 1998-2001 it is possible to see a reduction in SO_2 and NO_x in many of the most polluted sites, but the reduction is not so significant as in the first phase of programme. Together with this situation the decreasing of yearly corrosion loss of carbon steel and zinc in the last period had been close to previous exposure period.

Corrosion losses of six comparable one-year exposure periods evaluated within the programme show a decreasing trend.

The corrosion losses of carbon steel were reduced during period 1987 – 2001 on 20 test sites in Europe of 56% from average value 250 $g.m^{-2}$ to 140 $g.m^{-2}$. This decreasing of corrosion losses had been caused by decreasing of SO_2 concentration in air together with decreasing of acidity of precipitations.

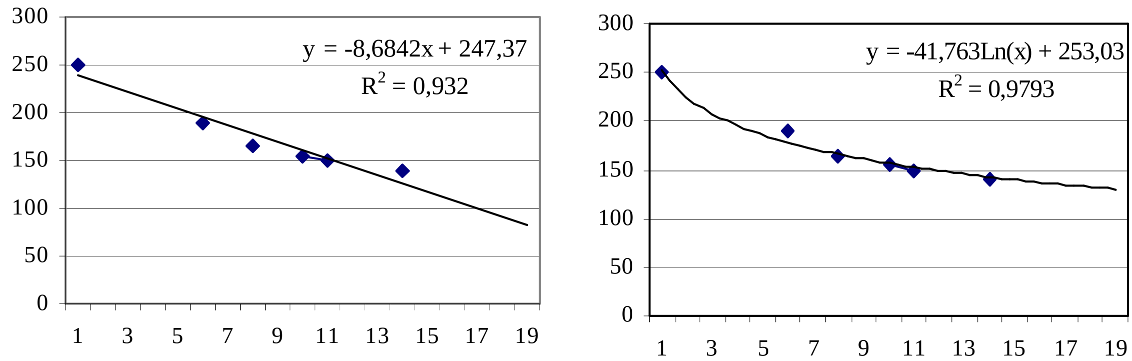
Figure 11 – Average corrosion loss of carbon steel on 20 test sites in Europe



The most significant decrease occurred at the end of 80ties. In last five years the pollution of SO_2 at majority of test sites did not change so dramatically as in previous decade. The corrosion losses of carbon steel in periods 1997/98 and 2000/01 had been stable on the similar level for the majority of test sites from original programme. In next years the trend of decreasing of corrosion loss of carbon steel would continue but probably in slow tendency. The best fitting of next progress looks to proceed according logarithmic trend (Figure 10). The concentration of SO_2 is on very lower level in majority test sites; the corrosion rate can be influenced significantly by other pollutants.

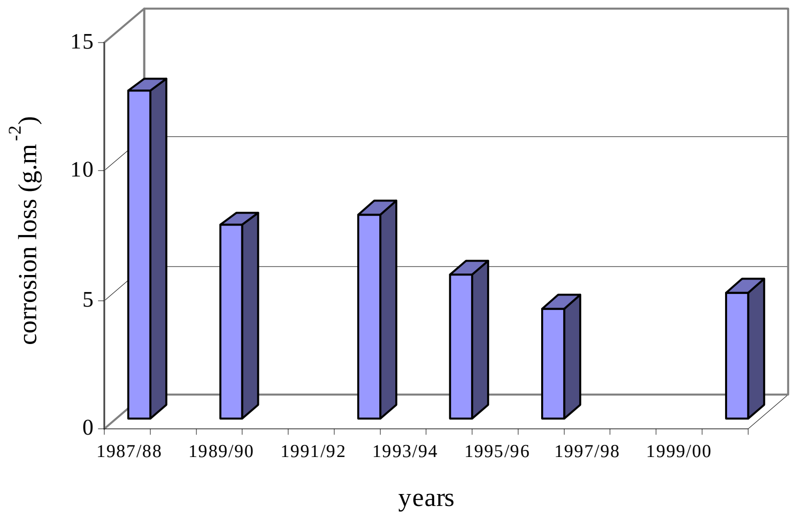
Figure 12 – Trend in decreasing of corrosion loss of carbon steel

linear logarithmic



The corrosion losses of zinc were reduced during period 1987 – 2001 on 20 test sites in Europe of 61% from average value 13 g.m^{-2} to 5 g.m^{-2}. Also this decreasing of corrosion losses had been caused by decreasing of SO_2 concentration in air together with decreasing of acidity of precipitation.

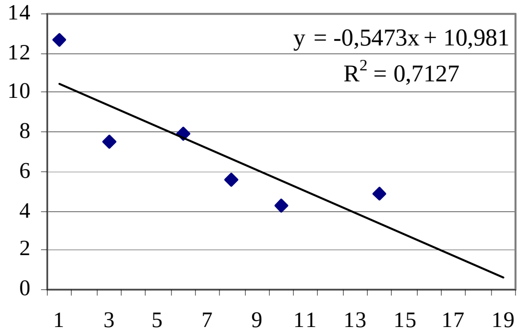
Figure 13 – Average corrosion loss of zinc on 20 test sites in Europe



The decreasing trend for corrosion loss of zinc is very similar to one for carbon steel. The best fitting of next progress looks to proceed according logarithmic trend too (Figure 12), but the value of R^2 in regression equations are lower than for carbon steel.

Figure 14 – Trend in decreasing of corrosion loss of zinc

linear



logarithmic

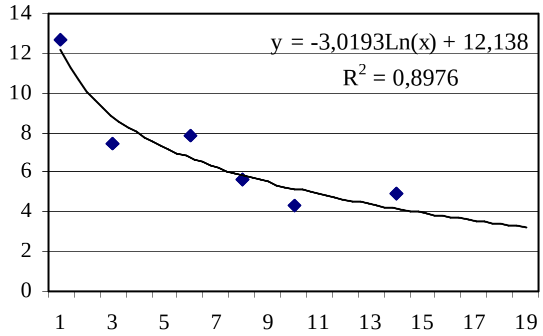
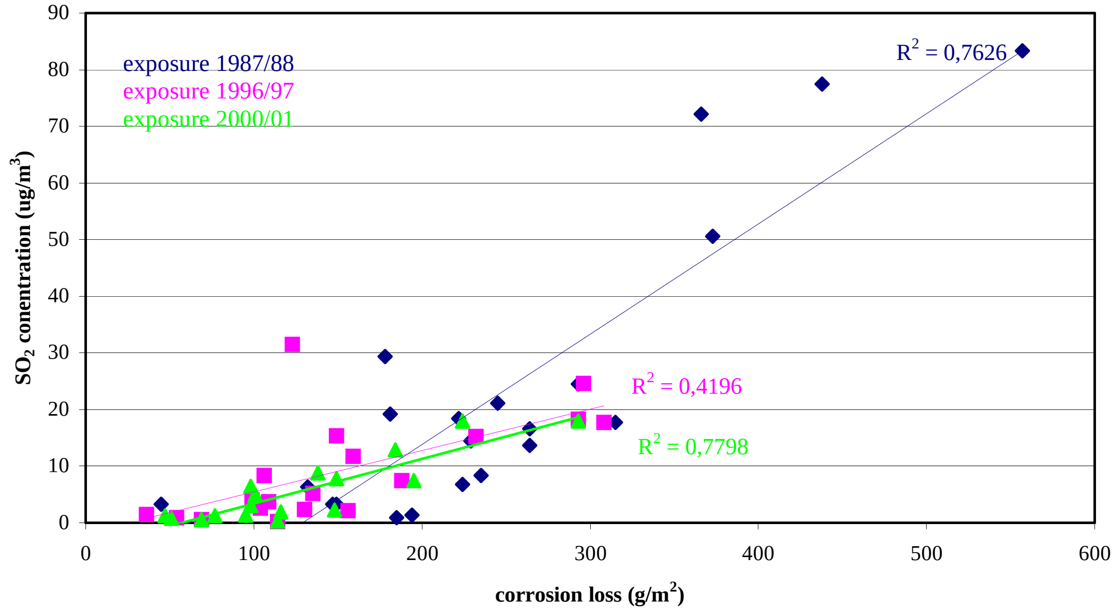


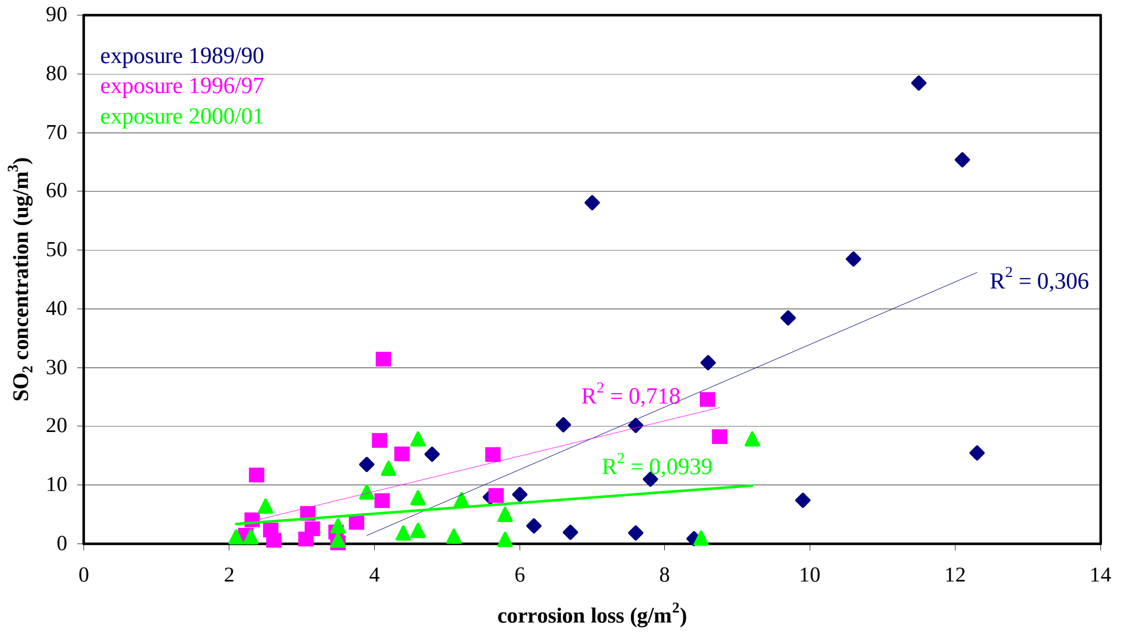
Figure 15 shows the relation between SO_2 concentration and corrosion loss of carbon steel and zinc. The last two periods are very similar for carbon steel, the R^2 value was much higher for period 2000/01. This value is low for period 1997/98 due to test site No 34. In case of zinc R^2 values are very low. In environments with SO_2 concentration below 10 g.m^{-3} this pollution is not he most significant factor for short-term corrosion losses of zinc. More significant for corrosion losses is relative humidity and time of wetness in such environments. Also in case of 1, 2 and 4 years exposure of zinc the corrosion losses after the 1st year had been significantly higher than after long-term exposure for 50% test sites [2].

Figure 15 – The relation between SO_2 concentration and corrosion losses of carbon steel and zinc

carbon steel



zinc



4. Conclusions

Results document the decreasing trend of pollution of SO_2 and also decreasing trend of corrosion loss of carbon steel and zinc in the period 1988-2001. Results show the efficiency of efforts for decreasing of pollution level in Europe.

Evaluation of trend samples for period 2000/01 included analysis of corrosion stimulators - anions - in corrosion products for samples exposed at test sites with high level of pollution. The runoff of zinc was determinated too.

Results will be evaluated deeper in connection with the next trend exposure, mainly on test sites included into programme since 1997. The complex environmental information will be considered.

Values of weight changes give a complementary information for trend samples only; they are important for runoff determination. These values are significant for other exposed materials in frame of this programme.

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Reference

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2. Faller M., Guseva O., Report No. 43 Results from Multipollutant programme: Corrosion attack on zinc after 1, 2 and 4 years of exposure (1997-2001), EMPA, Dubendorf, June 2003
3. Report No 41 Final Environmental data report for the multi pollutant programme: November 1997 to October 2001, 2003